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A production rate of 0.065 ± 0.003 Ar³⁷ atom/kg min of K³⁹ at 2-mwe depth below sea level was measured by sweeping argon from potassium solutions. This rate is unaffected by surrounding the solution by paraffin and is attributed to negative-muon captures and the electromagnetic interaction of fast muons, and not to a nucleonic cosmic-ray component. The Ar³⁷ yield from K³⁹ by the stopping of negative muons in a muon beam of the SREL synchrocyclotron was measured to be $8.5 \pm 1.7\%$. The stopping rate of negative cosmic-ray muons at 2-mwe depth below sea level from these measurements and an estimated 17% electromagnetic production is 0.63 ± 0.13 μ^- /kg min. Previous measurements on the muon stopping rate vary by a factor of 5. Our value is slightly higher but is consistent with two previous high values. The sensitivity of the Ar³⁷ radiochemical method for the detection of muons is considerably higher than that of the previous radiochemical methods and could be used to measure the negative-muon capture rates at greater depths.

Introduction

Until the experiments of the Utah cosmic-ray group [Bergeson et al., 1967], it was universally believed that cosmic-ray muons result solely from the decay of secondary pions and kaons from cosmic-ray interactions in the atmosphere. The Utah experiment raised the possibility for an additional source of muons. An explanation [Bjorken et al., 1969] of the Utah experiment has been proposed that requires an anomalous absorption of muons of negative helicity and unusual shape for the sea-level muon energy spectrum. This spectrum is often inferred from stopping rate versus depth measurements near sea level. Although the muon stopping rate has been

measured by three different methods, measurements by even the same method often disagree. Stopped muon rates have mainly been measured with nuclear emulsions and with scintillators at depths to 300 mwe (meters of water equivalent). Recent measurements by Bergamasco [1970] are a factor of 2-5 higher than previous ones. Additional measurements of the stopped negative-muon rate might be helpful in determining whether cosmic muons result solely from the decay of secondary π and κ mesons.

Until now, because of restricted sensitivity, radiochemical methods for detection have yielded positive results to a depth of only 0.2 mwe. Winsberg [1956] extracted Cl^{39} from rainwater produced by the $\text{Ar}^{40}(\mu^-, \nu_{\mu} n)$ reaction. Rama and Honda [1961] extracted gold radioactivities from mercury at sea level and state that their result is consistent with Winsberg's. Takagi and Tanaka [1969] observed a Fe^{59} radioactivity of 0.35 ± 0.14 atom/kg min induced by cosmic-ray muons in cobalt at sea level. This is a lower limit for the stopping of negative muons because the yield of the reaction was not determined. A more sensitive radiochemical method with a measured yield is needed.

The $\text{K}^{39}(\mu^-, \nu_{\mu} 2n)\text{Ar}^{37}$ reaction offers the possibility for extremely high sensitivity because it is possible to extract argon with almost 100% efficiency from large volumes of potassium solution and because Ar^{37} can be counted in small proportional counters with extremely low backgrounds. Davis et al. [1968] have employed a similar radiochemical method for the detection of neutrino-produced Ar^{37} from Cl^{37} in which he used volumes of chlorine solution 30,000 times larger than we have of potassium solution.

Experimental Procedures

Ar^{37} from cosmic-ray muons on potassium. The apparatus for the detection of cosmic-ray muons was located at sea level in our laboratory. Overhead building material contributed approximately 2 mwe of shielding. Figure 1 shows the apparatus, which consists of dissolution, gas-purification, and counter-filling sections. With the system closed off under a helium atmosphere, 6.78 kg of potassium acetate were

dissolved in a three-neck flask in 14.15 liters of distilled water. The argon content of the solution was then removed by flushing with purified helium. The helium entered the flask through a frit; a stirrer distributed the small helium bubbles throughout the solution. After repeated helium sweeps showed that less than 0.01 cc STP of argon remained in the solution, the flask was isolated for 72 days in order to allow for accumulation of the Ar^{37} induced by cosmic-ray muons. Then the solution was flushed again with helium and carrier argon (0.5 cc STP), which together removed the gases from the solution. The helium and carrier argon were then passed through a condenser and an alumina column to remove water vapor. All gases except helium and neon were then trapped on charcoal at -196° .

The gases trapped on charcoal were transferred to the purification section, where the chemically active constituents were gettered by titanium at 850°C . The remaining rare gases were trapped on charcoal at -196°C , and the argon was removed from the charcoal at -70°C . The argon yield was determined and together with 10% methane was transferred into a proportional counter of 0.5-cc volume of a design similar to that used by Davis et al. [1968]. The counting was done under low-level conditions; the low-level shield and a block diagram of the electronics are shown in Figure 2. Counting data for the 72-day exposure are shown in Figure 3.

The argon yield was 75% from the first sweep. By repeated helium sweeps, an additional 25% was obtained. This proved that the system was tight and the sweeps prepared the system for the next run. The solution was again isolated under a helium atmosphere and shielded by 10–15 cm of paraffin. After 48 days, the argon extraction and counting procedure was repeated. Table 1 summarizes the data for the two runs.

Ar^{37} from cyclotron muons on potassium. Reagent-grade potassium acetate, (CH_3COOK), was bombarded in a muon beam of the SREL synchrocyclotron. The target consisted of three 8-g samples of CH_3COOK that had been sealed under rough vacuum in quartz vials. The number of muons stopped in the CH_3COOK was measured to be $(3.25 \pm 0.63) \times 10^5/\text{g}$ by a counter telescope.* The number of muons stopped in

* Drs. K. Gotow and D. Bryman determined the number of stopped muons.

empty vials was subtracted from the number stopped in filled vials. The backward muon beam, which has a π -meson contamination of less than 1%, was used. Neutrons were monitored in the vicinity of the target. Figure 4 shows the experimental arrangement at the synchrocyclotron.

First, the gas contents of the quartz vials were removed with carrier argon (0.5 cc STP). The argon radioactivities in these gases were counted after purification. The potassium acetate and calcium samples were then either melted or dissolved, and their argon radioactivities measured. The argon extraction by melting was done in a manner similar to that we commonly use for meteorite measurements [Fireman and Goebel, 1970]. The argon extractions by dissolution were done in a manner similar to that described for the cosmic-ray muon experiment. Table 2 summarizes the target information and counting data.

Results

Cosmic-ray muon produced Ar³⁷. The counting data were reduced on a CDC 6400 computer. The errors given in Tables 1 and 2 include those from counting statistics (1 standard deviation) and estimates of other uncertainties. Figure 3 shows the counting data from a sweep of the potassium solution with 72 days of exposure time. The Ar³⁷ activity appears at a peak at 2.8 kev (channel 34) owing to the Auger electron from Ar³⁷ decay. The rate of growth of the Ar³⁷ peak with time is consistent with its 35-day half-life. The counting data from another sweep with 48 days of exposure time gave a slightly smaller peak at 2.8-kev energy. The counts in the neighborhood of channel 34 (2.8 kev) are more than 30 times the background rate of the counter for these channels. The counter background is continuously distributed, with no peaks, throughout all channels. Other rare-gas activities such as those of Ar³⁹ and Kr⁸⁵ are also continuously distributed throughout all channels. The continuously distributed activity in the argon from the solution flushes was three times the normal counter background. This excess, which was constant with time and raised the counter background, is probably due to atmospheric Kr⁸⁵ dissolved in the solution. Since the Ar³⁷ is clearly observable above the continuous activity, no effort was made to remove Kr⁸⁵ by gas chromatography. However, if the muon experiment were extended to great depths and much larger volumes of solution, it would be necessary to remove

the krypton by gas chromatography. Table 1 shows that equal amounts of Ar^{37} at saturation were produced in the potassium acetate solutions with and without the paraffin shielding; therefore, the fast-neutron contribution to Ar^{37} production in the solution from the nucleonic component of cosmic rays is negligible.

Cyclotron muon produced Ar^{37} . The Ar^{37} activities in two of three potassium acetate vials placed in the cyclotron negative-muon beam were measured. It is interesting to note (see Table 2) that all the Ar^{37} activity was in the gas phase of the potassium acetate vials and that none of the Ar^{37} from the calcium was in the gas phase. The gas phase of one acetate vial had 1.08 ± 0.06 dpm of Ar^{37} ; the other vial had 1.15 ± 0.08 dpm of Ar^{37} at the end of the bombardment. No Ar^{37} activity was observed when 1.4 g of potassium acetate were melted or 5.7 g dissolved. The Ar^{37} activity from the potassium acetate is attributed to muons stopped in K^{39} . The number of Ar^{37} atoms produced per 1 g of K^{39} is $(1.02 \pm 0.05) \times 10^4$. On the basis of the cyclotron counter telescope measurements and the weight percent of K^{39} in potassium acetate, the number of muons stopped per 1 g of K^{39} is $(1.20 \pm 0.23) \times 10^5$. The Ar^{37} yield from potassium acetate is $(1.02 \pm 0.05) \times 10^4 / (1.20 \pm 0.23) \times 10^5 = 8.5 \pm 1.7\%$. The Ar^{37} produced by muons in potassium acetate salt rapidly leaks into the gas phase at room temperature. This result is consistent with our not finding Ar^{37} in unsealed laboratory potassium acetate salt in a preliminary experiment.

The gas phase removed from the vial containing calcium metal had no Ar^{37} activity. Melting the calcium released 0.022 ± 0.005 dpm of Ar^{37} . This small activity from calcium indicates that the neutron flux in the neighborhood of the muon beam is negligible. However, approximately 0.07 dpm of Ar^{39} was observed in the Ca, probably due to negative muon captures in Ca^{40} . A determination of the yield of Ar^{39} from muon captures in Ca^{40} would be of interest to theorists in nuclear structure.

Number of stopped negative cosmic-ray muons. We have measured the Ar^{37} production in potassium acetate solution induced by cosmic-ray muons and the yield of Ar^{37} from potassium acetate in a negative-muon beam.

Tanaka et al. [1968] estimated the ratio of the Al^{26} produced by stopped muons and that by fast muons versus depth. They calculated that at sea level, approximately 17% of the muon-produced Al^{26} activity comes from electromagnetic interaction of fast muons. The two reactions $\text{Si}^{28}(\mu^-, \nu_{\mu} 2n)\text{Al}^{26}$ and $\text{Si}^{28}(\gamma, np)\text{Al}^{26}$ are similar to $\text{K}^{39}(\mu^-, \nu_{\mu} 2n)\text{Ar}^{37}$ and $\text{K}^{39}(\gamma, np)\text{Ar}^{37}$ reactions. The atomic numbers differ by only five units, so we use the 17% estimate of Tanaka et al. [1968] to correct for the electromagnetic interaction of fast muons.

Our measured Ar^{37} production rate of 0.065 ± 0.003 atom/kg min of K^{39} at 2-mwe depth reduced by 17% gives 0.054 ± 0.003 Ar^{37} atom/kg min of K^{39} produced by negative-muon captures. The Ar^{37} yield for negative-muon captures in K^{39} was measured to be $8.5 \pm 1.7\%$. The stopping rate of negative cosmic-ray muons at 2-mwe depth is $[(0.054 \pm 0.003)/(0.085 \pm 0.017)]$ or 0.63 ± 0.13 muon/kg min.

It should be mentioned that we used potassium acetate salt as a cyclotron muon target and potassium acetate solution for the cosmic-ray muon exposure. On the basis of the Fermi-Teller Z-law for muon captures, we estimate that this difference has a small effect on the capture rate. Most of our error is caused by the large error that we assigned to the cyclotron yield determination; this error could be reduced by further muon irradiations of the cyclotron. A cyclotron target consisting of a potassium acetate solution would be preferable to the target of potassium acetate.

Discussion

Table 3 gives our result, together with those of other investigators.

Three previous radiochemical experiments had been done. Rama and Honda [1961] found gold radioactivities from laboratory mercury and state that their result is consistent with a stopped-muon flux of $0.26 \mu^-$ stopped/kg min, a value that they derive from the experimental findings of Winsberg [1956]. Tanaka et al. [1968] found no muon-produced Al^{26} (7.4×10^5 yr) radioactivity in terrestrial silicates. A limit for the muon stopping rate from this experiment cannot be obtained without a knowledge of the yield. The $\text{Si}^{28}(\mu^-, \nu_{\mu} 2n)\text{Al}^{26}$ reaction may occur through the excited state

of Al^{26} , which has a 6.4-sec half-life. Takagi and Tanaka [1969] measured the muon-capture production rate of Fe^{59} radioactivity from cobalt at sea level to be 0.35 ± 0.14 atom/kg min. They calculate that the Fe^{59} from the (n, p) reaction on cobalt is negligible. Because some of the captured muons produce iron isotopes other than Fe^{59} , their measurement gives a lower limit of $(0.35 \pm 0.14)\mu^-$ stopped/kg min at sea level.

Three experiments with nuclear emulsions have been done. With strips of Ilford emulsions, Short [1963] measured the stopped negative-muon rate at 58-mwe depth underground. His stopping rate is 2.5 times higher than that obtained by George and Evans [1955] at 58-mwe depth with glass-backed nuclear emulsions. From his best estimate of the momentum spectrum of muons at sea level, Short [1963] also calculated the expected stopping rates at depths underground of 7-70 mwe. His stopping-rate - depth curve, which was normalized to his measured rate at 58-mwe depth, is also a factor of 2.5 higher than the rates observed by George and Evans [1955] and by Kaneka et al. [1955] at depths shallower than 58 mwe. Short's stopping-rate - depth curve gives 0.37 muon/kg min stopped at 7-mwe depth. With a ratio of 0.44 of negative-to-total stopped muons, Short's value gives $0.16 \mu^-$ /kg min at 7-mwe depth underground. Takagi and Tanaka [1969] calculated that the number of muons stopped at 2 mwe and that at 7 mwe are practically identical. Because of the uncertainties in the calculated extrapolation to 2-mwe depth, we give Short's rate to be $0.2 \mu^-$ /kg min stopped at 2 mwe and the rates of George and Evans [1955] and Kaneka et al. [1955] to be $0.1 \mu^-$ /kg min at 2 mwe with a 50% error in Table 3.

With a liquid scintillator, Bergamasco et al. [1970] measured the stopping rate of muons at depths underground of 60-300 mwe. At 60-mwe depth, the stopping rate of Bergamasco et al. [1970] is a factor of 2 higher than that of Short [1963]; a factor of 5 higher than that of George and Evans [1955]; and a factor of 2 higher than that of Barton and Slade [1965], which was also obtained with a liquid scintillator. If the scintillator results are extended to 2-mwe depth in the manner described, stopping rates of 0.4 and 0.2 negative muon/kg min at 2 mwe are obtained from Bergamasco et al. [1970] and from Barton and Slade [1965], respectively.

Our value of $0.63 \pm 0.13 \mu^-$ stopped/kg min is consistent with a value of $\geq 0.35 \pm 0.14 \mu^-$ stopped/kg min obtained by Takagi and Tanaka. If a 50% error is assigned to the extrapolation from 60 mwe to 2 mwe in depth, our value is consistent with that of Bergamasco et al. [1970] but not with those of Barton and Slade [1965], Short [1963], George and Evans [1955], and Kaneka et al. [1955]. According to Short's calculation, the stopping rate at 60-mwe depth underground is 1/28 of the stopping rate at sea level. Since the Ar^{37} activity observed from 6.78 kg of potassium acetate was 30 times the counter background, the Ar^{37} should be observable from 6.78 kg of potassium acetate at 60-mwe depth. Bergamasco et al. [1970] found the muon stopping rate at 300 mwe to be 1/10 the rate at 60 mwe; therefore, 70 kg of potassium acetate would be necessary to observe Ar^{37} from negative-muon captures at 300 mwe.

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TABLE 1. Data Summary for Potassium Acetate Solutions at 2-mwe Depth

Sample	Experiment	CH ₃ COOK Dissolved in Distilled H ₂ O	CH ₃ COOK Dissolved in Distilled H ₂ O Paraffin Shielded
Weight CH ₃ COOK (kg)		6.78	6.78
Distilled H ₂ O (kg)		14.15	14.15
Argon yield (%)		75 ± 2	97 ± 2
Days after extraction counted		0.5-13.0	1.0-25.0
Ar ³⁷ (cpm) [*]		0.060 ± 0.003	0.048 ± 0.002
Counting efficiency (%)		48.1 ± 2.0	50.0 ± 2.0
Ar ³⁷ (dpm) [*]		0.125 ± 0.007	0.096 ± 0.006
Days of exposure		72	48
Ar ³⁷ _{sat} (dpm/kg K ³⁹) [†]		0.066 ± 0.004	0.064 ± 0.004

* At time of extraction.

† Saturation activity.

TABLE 2. Data Summary of the Synchrocyclotron Negative Muon Bombardment

Sample	Calcium Metal Target		CH ₃ COOK [†] Target 1		CH ₃ COOK [†] Target 2	
	Total Gas from Vial	Calcium	Total Gas from Vial	CH ₃ COOK	Total Gas from Vial	CH ₃ COOK from Vial
Amount processed	with 4.0 g	4.0 g	with 8.0 g	1.4 g (melted)	5.7 g (dissolved)	with 8.0 g
Argon yield (%)	94 ± 1	98 ± 2	99 ± 1	48 ± 2	83 ± 2	88 ± 1
Days after EOB* counted	2-4	4-17	9-14	24-25	29-32	36-38
Ar ³⁷ _{EOB} (cpm)*	<0.005	0.010 ± 0.002	0.433 ± 0.010	<0.006	<0.004	0.517 ± 0.022
Counting efficiency (%)	45 ± 2	45 ± 2	40 ± 2	40 ± 2	40 ± 2 [†]	45 ± 2
Ar ³⁷ _{EOB} (dpm)*	<0.011	0.022 ± 0.005	1.08 ± 0.06	<0.015	<0.010	1.15 ± 0.08

* EOB = end of bombardment.

[†] Number of μ⁻ stopped/g (CH₃COOK) = (3.25 ± 0.63) × 10⁵.

TABLE 3. Comparison of Stopped Muon Rates Obtained by Various Investigators

Publication	Experimental Technique	Stopped μ^- /kg min	Depth (mwe = meters water equivalent underground)
This work	Ar ³⁷ from K ³⁹	0.63 ± 0.13	2
<u>Takagi and Tanaka</u> [1969]	Fe ⁵⁹ from Co ⁵⁹	≥0.35 ± 0.13	sea level
<u>Rama and Honda</u> [1961]	Au from Hg	(consistent with ~0.26)	sea level
<u>Short</u> [1963]	Nuclear emulsion	0.2	* 2 (Extrapolated from 58 mwe according to text)
<u>Kanaka et al.</u> [1955]	Nuclear emulsion	0.1	* 2 (Extrapolated from 17 mwe according to text)
<u>George and Evans</u> [1955]	Nuclear emulsion	0.1	* 2 (Extrapolated from 8 mwe according to text)
<u>Bergamasco et al.</u> [1970]	Liquid scintillator	0.4	* 2 (Extrapolated from 60 mwe according to text)
<u>Barton and Slade</u> [1965]	Liquid scintillator	0.2	* 2 (Extrapolated from 60 mwe according to text)

* The fraction of stopped muons that are negatively charged is taken to be 0.44.

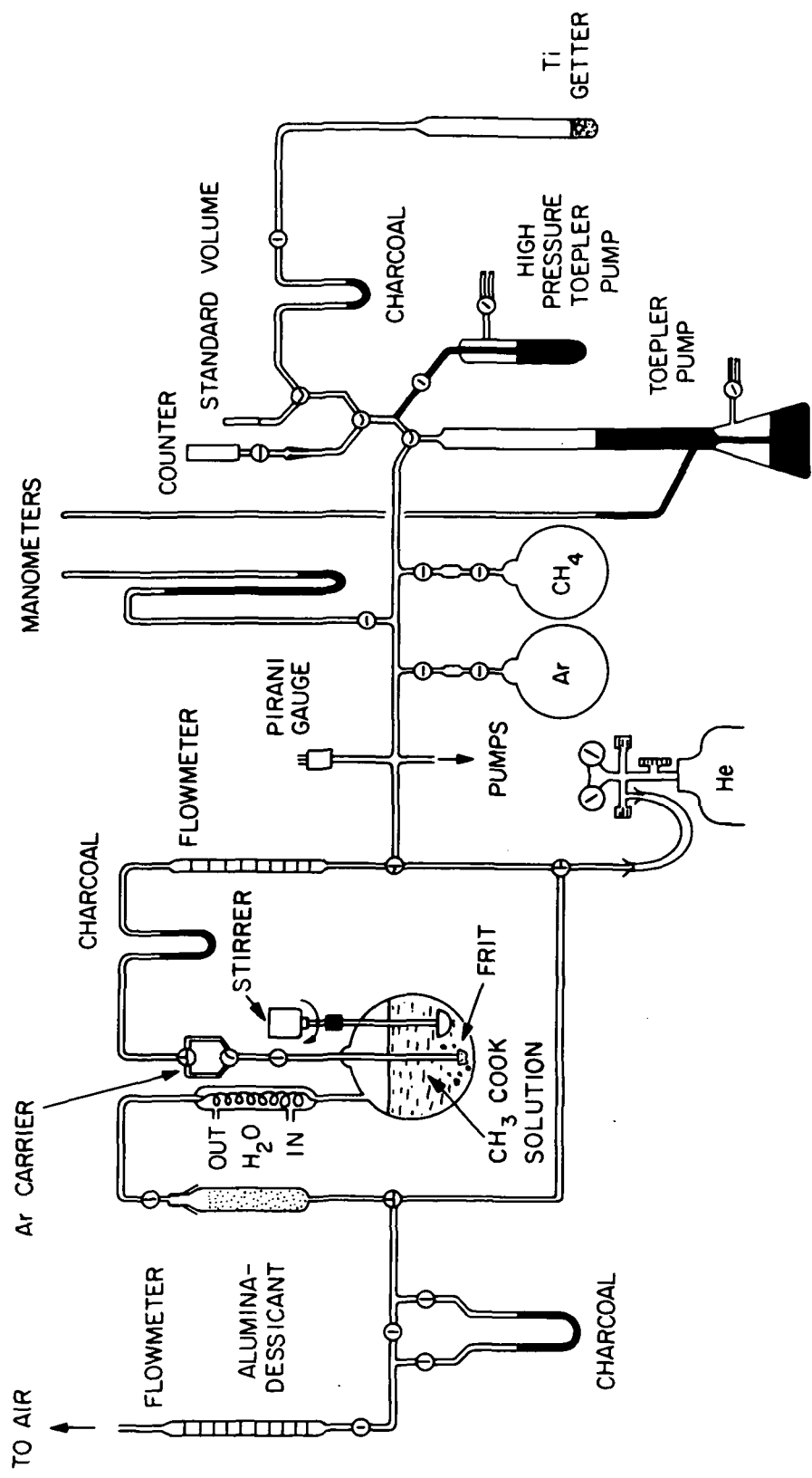


Fig. 1. System for the bombardment of potassium acetate solution with cosmic-ray muons. The gas - extraction and purification sections and the counterfilling arrangement are also shown.

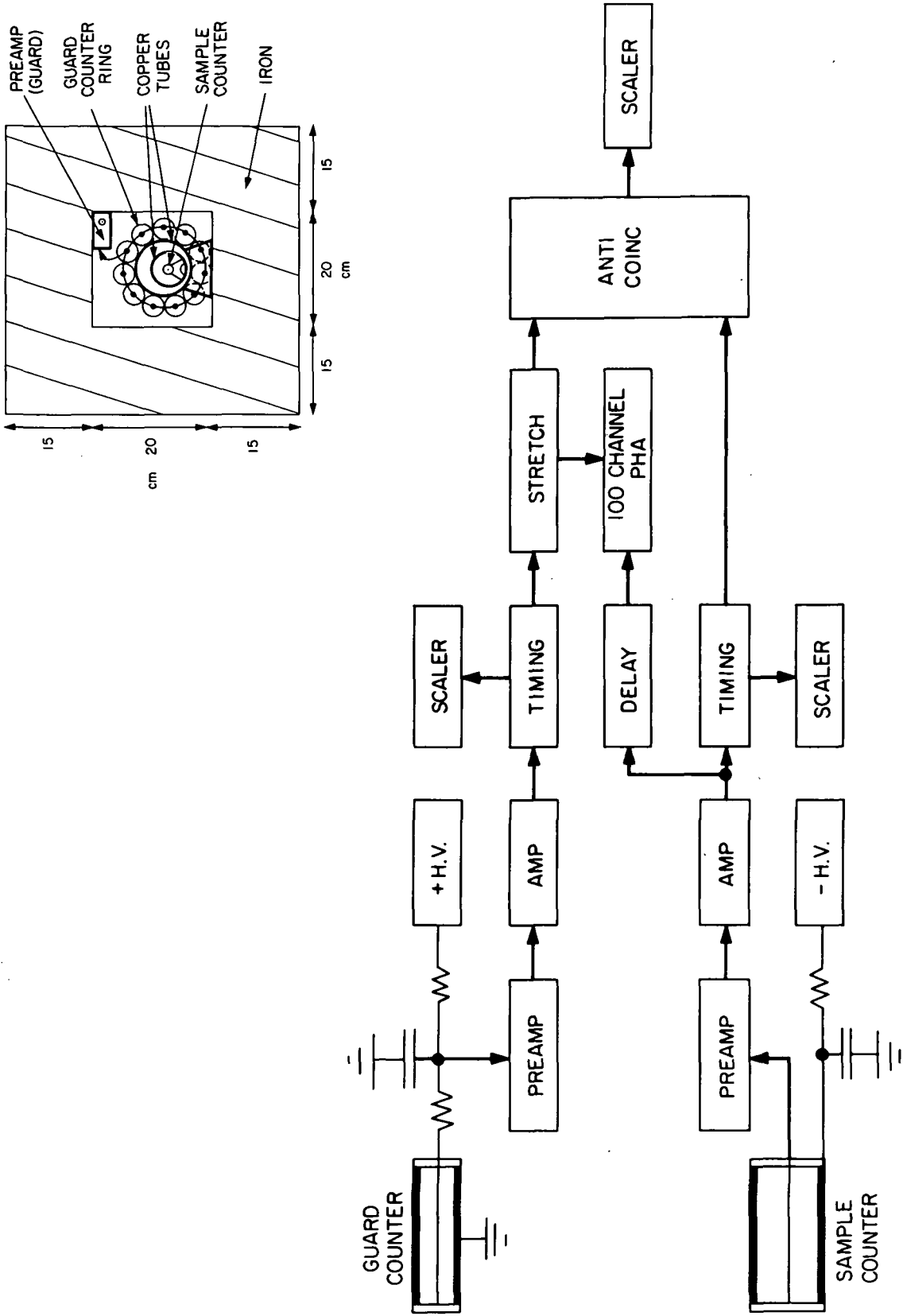


Fig. 2. Low-level shield and block diagram of electronics.

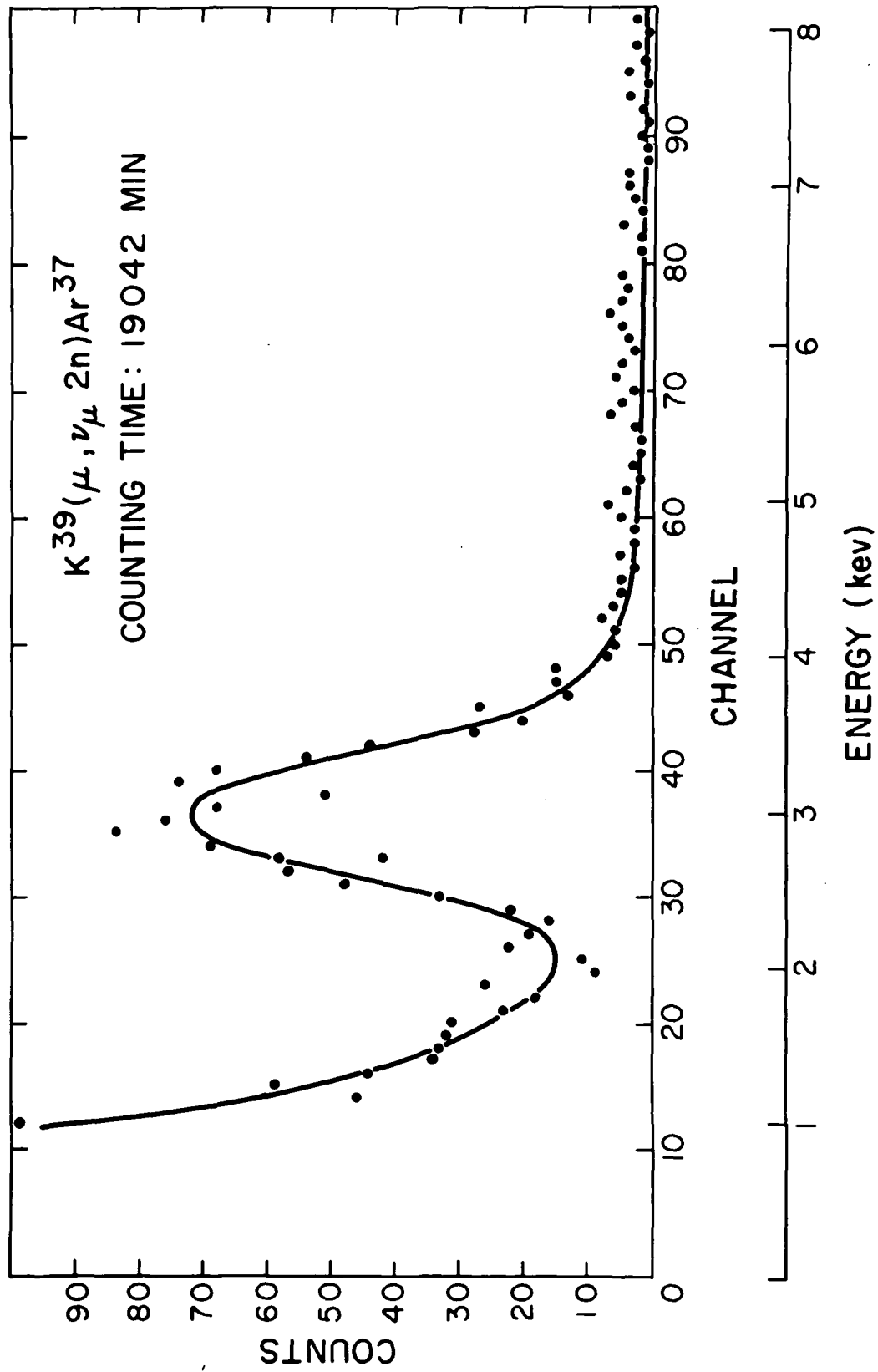


Fig. 3. Spectrum of the Ar^{37} radioactivity that was extracted from the cosmic-ray muon-bombarded potassium acetate solution (experiment 1).

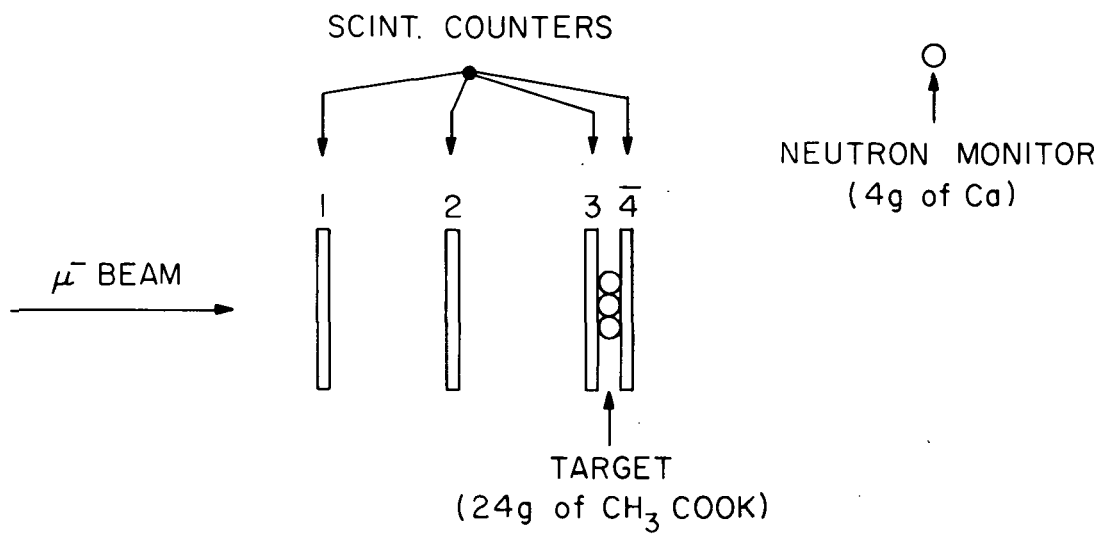


Fig. 4. Experimental setup at the SREL synchrotron.

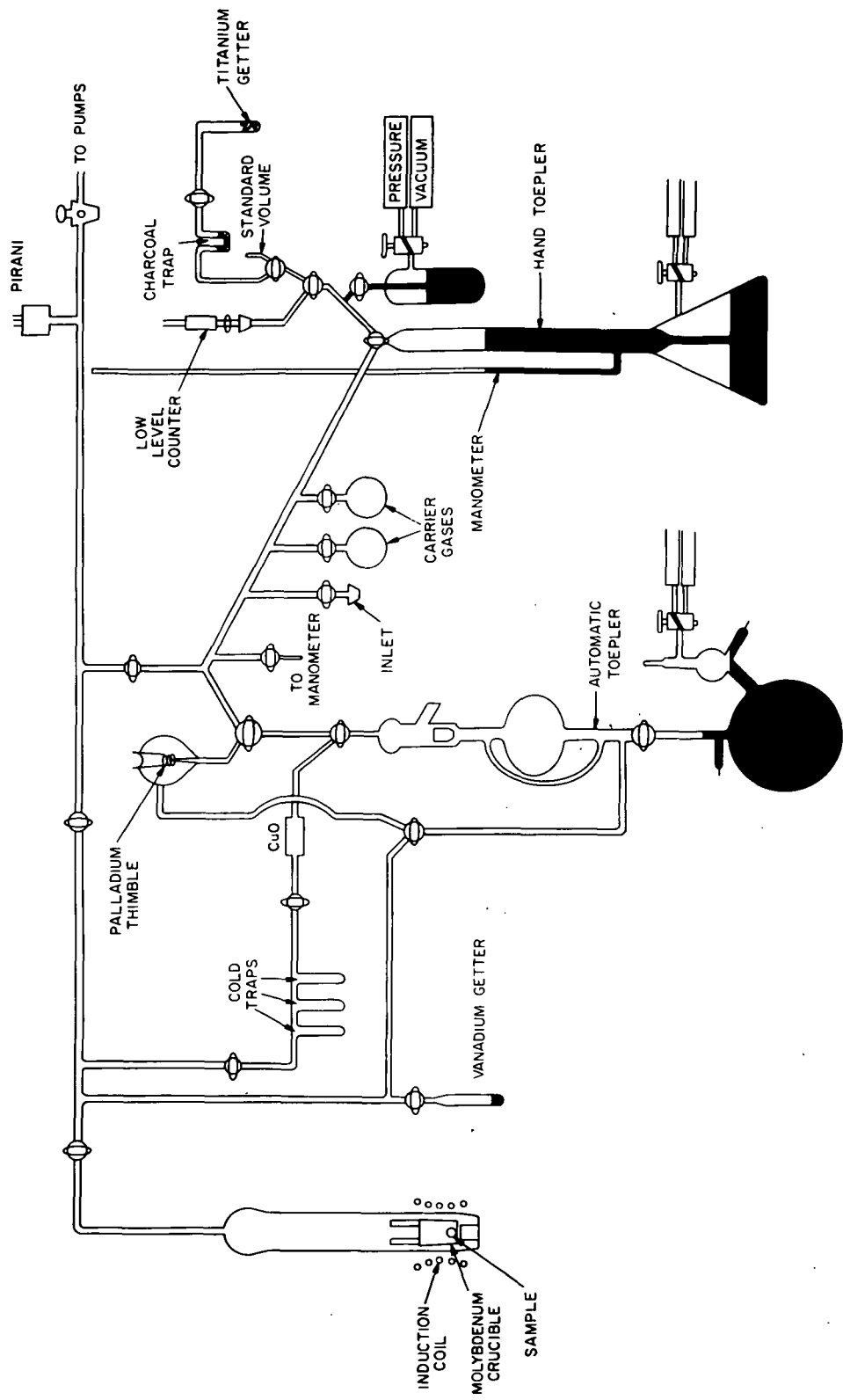


Fig. 5. Apparatus for melting of samples and processing of the released gas.