

## A Search for Neutrinos from the Sun\*

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Abstract

A solar neutrino detector has been built that is based upon the  $\text{Cl}^{37}(\nu, e^-)\text{Ar}^{37}$  reaction. The detector uses 3900,000 liters of tetrachloroethylene as the neutrino capturing medium. Argon is removed from the liquid by sweeping with helium gas, and counted in a small low level proportional counter. The recovery efficiency of the system was tested with  $\text{Ar}^{36}$  by the isotope dilution method, and also with  $\text{Ar}^{37}$  produced in the liquid by fast neutrons. These tests demonstrate that  $\text{Ar}^{37}$  produced in the liquid by neutrino capture can be removed with a 95 percent efficiency by the procedure used.

Initial results with the detector show that the neutrino capture rate was less than 0.5 per day, corresponding to a total neutrino flux-cross section product of less than  $3 \times 10^{-36} \text{ sec}^{-1}$ . From this limit and the cross sections of Bahcall, the following conclusions were drawn: (1) the flux of neutrinos from  $\text{B}^8$  decay in the sun was less than  $2 \times 10^6 \text{ cm}^{-2} \text{ sec}^{-1}$ ; (2) less than 9 percent of the sun's energy is produced by the CNO-cycle; and (3) the extraterrestrial flux of 1, 10, and 100 MeV neutrinos is less than  $5 \times 10^9$ ,  $13 \times 10^5$ , and  $4 \times 10^2 \text{ cm}^{-2} \text{ sec}^{-1}$ , respectively.

The ultimate detection sensitivity of the present experiment is limited by background effects from cosmic radiation and internal contamination with natural alpha emitters. Knowledge of the magnitude of these background effects show that the ultimate neutrino detection sensitivity can be increased five-fold. A brief survey is given of possible low threshold radio-chemical neutrino detection techniques that could be used for observing low energy neutrinos from the sun.

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## A Search for Neutrinos from the Sun\*

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### Introduction

A solar neutrino detection system has been built to observe the neutrino radiation from the sun. Neutrino detection depends upon observing  $\text{Ar}^{37}$  produced in 610 tons of tetrachloroethylene ( $\text{C}_2\text{Cl}_4$ ) by the neutrino capture reaction  $\text{Cl}^{37}(\nu, e^-)\text{Ar}^{37}$ . The apparatus was completed in early 1967, and initial measurements were performed. Argon-37 was not observed in these experiments, and from the sensitivity of the counting measurements it was only possible to set an upper limit to the solar neutrino flux.<sup>1</sup> The total neutrino capture rate in  $\text{Cl}^{37}$  was found to be less than or equal to  $3 \times 10^{-36} \text{ sec}^{-1}$ . Of particular interest was the flux of neutrinos from the decay of  $\text{B}^8$  in the sun. The upper limit to the flux of  $\text{B}^8$  neutrinos was  $2 \times 10^6 \text{ cm}^{-2} \text{ sec}^{-1}$ , approximately a factor of seven below the flux predicted at that time by solar model calculations.<sup>2,3</sup> However, there became available a new measurement of the carbon, nitrogen and oxygen composition of the sun, and some additional nuclear data. When this new information was introduced into the solar model calculation the predicted  $\text{B}^8$  flux was reduced by a factor of three.<sup>4</sup> The present status of these solar model calculations will be discussed at this conference by Dr. Bahcall. A summary of his results is given in Table I. This table lists the fluxes of neutrinos from various processes occurring in the sun, and the corresponding neutrino capture cross sections in  $\text{Cl}^{37}$ . These calculations predict a total neutrino capture rate in  $\text{Cl}^{37}$  of  $0.6 \times 10^{-35} \text{ sec}^{-1}$ . It is interesting to note that

80 percent of this rate would arise from  $B^8$  neutrinos. This is a result of the high neutrino capture cross section that leads to the formation of the analog state in  $Ar^{37}$ , an important feature of the  $Cl^{37}(\nu, e^-)Ar^{37}$  reaction that was first pointed out by Dr. Bahcall.<sup>5</sup>

It has been suggested by Ezer and Cameron<sup>6</sup> that circulation in the sun associated with a fast rotating core could maintain a high hydrogen abundance in the interior of the sun, and as a consequence reduce the flux of  $B^8$  neutrinos. They estimated the maximum reduction in the  $B^8$  flux by this process, and found the flux to be 4.7 times lower than one would expect if the sun were not mixed. Some additional calculations of the effect of mixing in the sun have been made,<sup>7,8</sup> but at the present time it is not clear whether this process is important in the sun.

The present report will be devoted to the design, operation, various tests of the recovery of  $Ar^{37}$ , and the counting procedures that are used in the experiment. Some new results will be given that serve to substantiate the earlier results. A discussion will be given of possible improvements in the detection sensitivity of the experiment, and the limitations imposed by various background processes.

#### Design of the Detector

Solar neutrino detection by the  $Cl^{37}-Ar^{37}$  method depends upon the formation of a few hundred (or less) atoms of radioactive  $Ar^{37}$  by neutrino capture in a large volume of liquid tetrachloroethylene,  $C_2Cl_4$ . These few hundred atoms must be removed from the liquid with high efficiency, and placed in a small low level proportional counter to observe the characteristic radiations from the decay of  $Ar^{37}$ . These general ideas, and the advantages of the  $Cl^{37}-Ar^{37}$  method were first pointed out by Professor Pontecorvo 20 years ago.

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Since the solar neutrino flux is very low, it is necessary to use as large a volume of tetrachloroethylene as is technically feasible, and to perform the counting measurements in a miniature counter with a very low background counting rate. It is important to have a sealed system to prevent the inleakage of air argon, since it would dilute the final sample that is counted. To insure that the tank and liquid pumping system was absolutely tight, a helium leak test was performed. For this test the entire system was evacuated, the outside of the system was blanketed with helium gas, and a search for helium leaking into the system was made with a mass spectrometer attached to the vacuum pumping system. The total helium leakage rate was less than  $10^{-6} \text{ cm}^3 \text{ sec}^{-1}$  under these conditions. This test demonstrated that the inleakage of air argon into the system during the long exposures (100 days) would be small. Following this test the tank was filled with tetrachloroethylene.

The detector employs  $3.8 \times 10^5$  liters of tetrachloroethylene that was contained in a horizontal cylindrical tank 6.2 meters in diameter and 14.8 meters long. The  $\text{Ar}^{37}$  formed in the liquid by neutrino capture is removed by purging the liquid with helium gas. To provide the gas and liquid circulation a system of pumps and eductors are used, see Figure 1.

Liquid is pumped from the bottom of the tank, and returned through two header pipes that run longitudinally through the center of the tank. On each of these header pipes is attached a set of 20 equally spaced eductor (aspirator) nozzles that draw helium from the top of the tank and mix it with the liquid as fine bubbles. This system of pumps and eductors aspirates the helium blanket gas (volume 20,000 liters) through the liquid at a total rate of 9000 liters per minute. The combined agitation and bubbling action produces turbulent mixing of helium gas throughout the entire volume of the liquid. This provides an effective purging action to bring argon dissolved

in the liquid into the helium gas phase.

Helium in the tank is circulated through an argon extraction system and returned to the tank. Again, flow is maintained by a pair of eductors operating from the liquid pumping system. These provide a helium flow rate through the argon extraction system of 310 liters per minute. The components of the argon extraction are shown schematically in Figure 1. Helium leaving the tank first passes through a baffled condenser at  $-40^{\circ}\text{C}$  to condense the bulk of tetrachloroethylene vapors. The remaining tetrachloroethylene or any trace amounts of water that may be present are removed by a molecular sieve (dehydrated zeolite) absorber. Argon is finally removed from the helium by a charcoal absorber held at liquid nitrogen temperature ( $77^{\circ}\text{K}$ ), and then the helium is returned to the tank.

The apparatus was built 1490 meters underground (approx. 4400 m.w.e.) at the Homestake Gold Mine at Lead, South Dakota, U.S.A. The arrangement of the tank, pumps and argon extraction system in the mine is shown in Figure 2. The tank was set below the access tunnel, and a water tight door was provided between the tank chamber and the pump room. This arrangement permits flooding the tank chamber with water to form a fast neutron shield around the tank. Background effects from cosmic ray muons, fast neutrons and internal natural alpha emitters will be discussed later.

#### Ar<sup>37</sup> Recovery Efficiency Tests

The recovery efficiency of the system for removing argon from the liquid was tested by two methods. A measured volume of isotopically pure Ar<sup>36</sup> was placed in the tank by introducing it into the helium gas stream at the point shown in Figure 1. It was then dissolved in the liquid by operating the pumps and eductors. The Ar<sup>36</sup> was recovered from the tank by operating the system at the fixed helium gas flow rates stated earlier.

It was found that the volume of  $\text{Ar}^{36}$  in the tank dropped exponentially with the volume of helium circulated through the argon recovery system. A 95 percent recovery of the  $\text{Ar}^{36}$  introduced could be achieved by circulating 0.42 million liters of helium through the argon extraction system, and this can be accomplished in a period of 22 hours.

A second method of testing the efficiency for recovering argon from the tank was made using radioactive  $\text{Ar}^{37}$ . A Ra-Be neutron source was placed in a re-entrant iron pipe that extends to the center of the tank, see Figure 1. Fast neutrons with energy above 1.08 MeV produce  $\text{Ar}^{37}$  in the liquid by the two nuclear reactions,  $\text{Cl}^{35}(\text{n,p})\text{S}^{35}$  and  $\text{Cl}^{37}(\text{p,n})\text{Ar}^{37}$ . The  $\text{Ar}^{37}$  produced by the neutron irradiation was removed along with carrier  $\text{Ar}^{36}$  (introduced after the irradiation) by three successive helium purges. The recoveries of  $\text{Ar}^{36}$  and  $\text{Ar}^{37}$  are given in Table II. It may be observed that the recovery of  $\text{Ar}^{37}$  with successive helium purges does follow the recovery of  $\text{Ar}^{36}$  carrier. These tests are of crucial importance, and do demonstrate that  $\text{Ar}^{37}$  produced in the tank by neutrino capture would be removed with an efficiency of at least 95 percent by the helium purge.

#### Argon-37 Counting

The entire argon sample is purified by gas phase chromatography and placed in a small low level proportional counter. A rare gas separation is required to remove Kr and Xe that are present in the rare gases recovered from the tank. These higher rare gases were absorbed in the tetrachloroethylene from the atmosphere during manufacture and storage. The rare gases recovered from <sup>the</sup> tank, therefore, contains 10.8 year  $\text{Kr}^{85}$ , and this activity must be completely removed from the final argon sample that is counted. To ensure the complete removal of the higher rare gases a gas phase chromatographic separation is carried out through a charcoal column.



The counter has an internal volume of  $0.65 \text{ cm}^3$  and the active volume is  $0.49 \text{ cm}$  in diameter and  $3.0 \text{ cm}$  long. It is made of materials known to be free of radioactive contaminants. The cathode is made of zone refined iron, and the outer envelope is made of silica glass. The center wire is made of tungsten and is  $25 \text{ microns}$  in diameter. Since the materials used are metals and silica glass, the counter can be outgassed by heating, and, as a consequence it does not exhibit memory effects after counting relatively high level  $\text{Ar}^{37}$  samples. A window is provided at the end of the counter so that X-rays from an  $\text{Fe}^{55}$  source can be used for energy calibration, see Figure 3. The sample of argon gas along with 5 to 10 percent methane is introduced into the counter through two silica glass tubes with stopcocks that join to a common taper joint. The argon is compressed into the counter by a Toepler pump, and the mercury columns are brought to the ends of the capillary tubes that lead to the active volume of the counter.

The counter is operated inside the well of a  $12.7$  by  $12.7 \text{ cm}$  sodium iodide scintillation counter. Exterior to this is a mercury shield  $3 \text{ cm}$  thick, a ring of proportional counters, and a  $30 \text{ cm}$  thick iron shield. Events in anticoincidence with the scintillation counter and the ring of proportional counters are measured on a 100 channel pulse height analyzer, and recorded on a continuously advancing paper tape. The pulse shape is displayed on a storage oscilloscope and recorded photographically. In this way possible noise pulses can be noted and eliminated. Less than a few percent of the observed pulses are caused by electrical noise, and these are usually below channel 20. The voltage on the counter is set so that the  $2.8 \text{ keV}$  peak from the  $\text{Ar}^{37}$  decay is centered at channel 50 in the spectrum. The resolution of the counter for  $\text{Ar}^{37}$  is 28 percent (full width at half maximum), and, for events within this resolution, the counter has an efficiency of 51 percent.

## Results

Two experimental runs are here reported. One of these runs was reported earlier,<sup>1</sup> and was from a 110 day exposure from day 174 to day 284, 1967 (run no. 9). The recovery of Ar<sup>36</sup> carrier gas for this experiment was 95 percent. The pulse height spectra for a 35 day count and a 71 day count (started on day 177) are shown in Figure 3. These spectra may be compared to the background spectrum obtained with the counter filled with Ar<sup>36</sup> that was purified by the same procedure as used for the sample from the tank. It may be noted that 10 counts were observed in the argon sample recovered from the tank in the 14 channels centered about channel 50, whereas the background for an equivalent period of time was 11 counts. Hence, in this experiment (run no. 9) the difference in the total accumulated counts in the argon recovered from the tank and the background was  $-1 \pm 5$  counts for the 35 day period. There is no evidence of Ar<sup>37</sup> activity in the argon recovered from the tank, and the experiment can only give an upper limit to the Ar<sup>37</sup> production rate in the tank from solar neutrinos. Using 5 counts as an upper limit to the Ar<sup>37</sup> activity observed during the 35 day period, and allowing for radioactive growth and decay, argon recovery efficiency, and counting efficiency, one would conclude the Ar<sup>37</sup> production rate in the tank was less than or equal to 0.5 Ar<sup>37</sup> atoms per day.

Another experiment was performed in which the period of irradiation was 130 days from day 22 to 152, 1968 (run no. 12). An Ar<sup>36</sup> recovery efficiency of 97 percent was achieved, and the argon gas sample recovered had a volume of 0.42 cm<sup>3</sup>. The counting data for this experiment is given in Figure 4, along with a background count made immediately prior to introducing the argon recovered from the tank. The count was continued for 131 days, and the figure shows four sequential pulse height spectra obtained during this period.

The last period of counting can be used as a background count since only 10 percent of the initial  $\text{Ar}^{37}$ , if present, would remain. The chronological occurrence of each pulse during this period is shown in Figure 5, and it may be noticed that the counts occurred linearly with time, but with a large statistical variation because of the small number of counts. A comparison can now be made of the counts observed within the half-width of the  $\text{Ar}^{37}$  peak position from the sample of argon from the tank, the initial background count, and the residual background count. These are: 11 counts for the sample from the tank, 8.6 counts for the initial background, and 11.6 counts for the residual background, scaled to a 33.6 day period. The counts observed for the intermediate periods are somewhat lower, 8 and 3.5 counts scaled again for the 33.6 day period.

The initial background count appropriately combined with the residual background count can be used to determine the background counting rate for the counter, namely  $10.2 \pm 2.3$  counts in 33.6 days. Comparing this to the first 33.6 day count with the argon recovered from the tank in which 11 counts were observed, one can conclude that  $0.8 \pm 4$  counts can be contributed by  $\text{Ar}^{37}$ . This experiment (run no. 12) is entirely consistent with the previously discussed experiment (run no. 9). The number of counts observed during the first counting period, and the background counting rates were essentially identical.

It may be concluded from these two experiments that the  $\text{Ar}^{37}$  production rate, or solar neutrino capture rate in the  $3.8 \times 10^5$  liters of tetrachloroethylene is less than or equal to 0.5 per day, and therefore, neutrino capture rate per  $\text{Cl}^{37}$  atom  $\leq 3 \times 10^{-36} \text{ sec}^{-1}$ .

## Discussion

The upper limit of the neutrino capture rate per  $\text{Cl}^{37}$  atom is a factor of two lower than expected from current solar model calculations. The calculations of Dr. Bahcall and his co-workers in Table I, that give a total neutrino capture rate of  $6.2 \times 10^{-36} \text{ sec}^{-1}$ , were made using the accepted values of the various parameters introduced in the model. One can adopt the view, as Dr. Bahcall has done, that errors in the parameters are sufficiently large to accommodate the present result. To resolve this question would require an increase in the sensitivity of the experiment. For example, if the sensitivity of the experiment were increased by a factor of five, the combined low energy neutrino fluxes from  $\text{Be}^7$  and the  $\text{H}(\text{He}, \nu)\text{D}$  reaction should be observed. Perhaps an improvement in sensitivity by a factor of five is possible. The experimental approaches toward reaching this goal will be discussed later. Bahcall, Bahcall, and Shaviv<sup>4</sup> have shown that the flux of more energetic neutrinos (1.44 MeV) from the  $\text{H}(\text{He}, \nu)\text{D}$  reaction does not depend upon the parameters used in the calculation. Hence, if the sun is composed mainly of hydrogen, and is producing energy by thermal fission, a flux of 1.44 MeV neutrinos equal to  $1.7 \times 10^8 \text{ cm}^{-2} \text{ sec}^{-1}$  should be observable at the earth. This flux would give a capture rate of  $0.3 \times 10^{-36} \text{ sec}^{-1}$  per  $\text{Cl}^{37}$  atom. To observe these neutrinos alone would require an increase in sensitivity of more than a factor of ten over that achieved in the experiments reported here. It appears from our knowledge of background effects, that it is unlikely that the sensitivity of the present experiment will be increased enough to measure the neutrinos from the  $\text{H}(\text{He}, \nu)\text{D}$  reaction if this is the only source of solar neutrinos.

Since the cross section for the  $\text{Cl}^{37}(\nu, e^-)\text{Ar}^{37}$  reaction is well known,<sup>5,9,10,11</sup> the experimental limit on the capture rate can be used to calculate an upper limit on the flux of neutrinos from various specific

processes. In particular one can conclude the flux of  $B^8$  neutrinos from the sun is less <sup>than</sup> or equal to  $2 \times 10^6 \text{ cm}^{-2} \text{ sec}^{-1}$ . Also, if the CNO cycle is dominant in the sun, the flux-cross section product is  $35 \times 10^{-36} \text{ sec}^{-1}$ . This value<sup>5</sup> is determined by knowing the fraction of the energy radiated as neutrinos in the CNO cycle, the solar constant, and the cross section for neutrinos from  $N^{13}$  and  $O^{15}$ . One may then conclude from the experimental limit, that less than 9 percent of the sun's energy is produced by the CNO cycle. This conclusion is in agreement with current solar model calculations. For example, the fluxes given in Table I would infer that 0.7 percent of the sun's energy is produced by the CNO cycle.

The limit on the neutrino capture rate in  $Cl^{37}$  may also be used to set limits on the total extra-terrestrial electron neutrino flux. This is of interest for various cosmological considerations.<sup>12</sup> Table III summarizes the limits on the flux for neutrinos of energy 1, 5, 10, and 100 MeV energy, and the corresponding limits on the energy density of neutrino radiation. The limits are not very low at low energy, but at 100 MeV the energy density is approaching the range of that observed for total cosmic radiation, that is  $1 \text{ eV cm}^{-3}$ . Above a few hundred MeV the cross section will be affected by nucleon loss from the resulting  $Ar^{37}$  nucleus, and therefore the cross section is not well known.

#### Improvements in Sensitivity

The apparatus as described here was designed to measure a solar neutrino capture rate of  $30 \times 10^{-36} \text{ sec}^{-1} \text{ Cl}^{37} \text{ atom}^{-1}$ , or in the event that the solar neutrino flux is lower than expected the apparatus would be sufficiently sensitive to be able to search a factor of ten below this rate. These initial aims have been achieved, and one may now ask whether the sensitivity of the apparatus can be increased. From the foregoing discussion

it is clear that an improvement in sensitivity by a factor of five would be required to observe neutrinos from  $\text{Be}^7$  decay in the sun, and an increase in sensitivity of over ten is required to observe neutrinos from the  $\text{H}(\text{He}, \nu)\text{D}$  reaction if this is the only source of solar neutrinos.

The sensitivity of the present experiment is limited by various background processes that produce  $\text{Ar}^{37}$  in the tank. The most serious background effect is from cosmic ray muons. Protons produced by cosmic ray muon interactions form  $\text{Ar}^{37}$  in the liquid by the  $\text{Cl}^{37}(\text{p}, \text{n})\text{Ar}^{37}$  reaction. The  $\text{Ar}^{37}$  production rate from this process as a function of the depth underground was estimated from measurements performed at 25 m.w.e., the decrease in the muon intensity with depth, and the increase in the cross section for nuclear interaction of muons with depth. This rather crude analysis indicated the cosmic ray muon production of  $\text{Ar}^{37}$  at a depth of 4400 m.w.e. is 0.1 atom per day.<sup>13</sup> The study of the production of neutrons by nuclear interactions of muons underground of Ryajslcaya and Zatsepina<sup>14</sup> would indicate the background at this depth would be 0.06  $\text{Ar}^{37}$  atom per day. Professor Wolfendale<sup>15</sup> kindly estimated the background effect from fast muon interactions, and found also an  $\text{Ar}^{37}$  production rate of 0.06  $\text{day}^{-1}$ . These analyses are based upon an  $\text{Ar}^{37}$  production rate of 6500  $\text{day}^{-1}$  in  $3.8 \times 10^5$  liters tetrachloroethylene at a depth of 25 m.w.e., and they assume the muon interactions vary with depth as  $I\bar{E}^{-0.7}$ , where  $I$  is the total muon intensity and  $\bar{E}$  is the average muon energy. In view of the long extrapolation from the measurements at 25 m.w.e. to a depth of 4400 m.w.e., the estimated muon background effect is not very accurate. Experiments are in progress with a 16,000 liter detector that may be set at various depths in the mine. A single measurement was performed at a depth of approximately 800 m.w.e., and the  $\text{Ar}^{37}$  production rate was found to be less

than 0.4 atoms per day. Whereas the above analysis would predict a rate of 1.3 Ar<sup>37</sup> atoms per day in 16,000 liters of tetrachloroethylene. Additional measurements will be performed to derive a more accurate value for the muon background effect in the 380,000 liter detector. However, for the present we must use a rate of 0.06 Ar<sup>37</sup> atom per day as the best estimate of the cosmic ray background. The upper limit on the Ar<sup>37</sup> production rate of 0.5 atom per day upon which the limit for the solar neutrino flux is based, is then a factor of 8 above the cosmic ray background.

The next most serious background effect arises from fast neutrons (energy above 1 MeV) produced by spontaneous fission of uranium and ( $\alpha$ ,n) reactions in the surrounding rock wall. These neutrons penetrate the wall of the tank producing protons by the Cl<sup>35</sup>(n,p)S<sup>35</sup> reaction, which then form Ar<sup>37</sup> by the Cl<sup>37</sup>(p,n)Ar<sup>37</sup> reaction. The rock in the tank room is low in uranium and thorium. Measurements of various samples of the rock by gamma scintillation counting gave the following compositions: uranium 0.2 to 5 ppm and thorium 1.3 to 24 ppm. The magnitude of this fast neutron background effect has been measured with a radiochemical fast neutron detector. Neutron detection depends upon observing Ar<sup>37</sup> produced by the Ca(n, $\alpha$ )Ar<sup>37</sup> reaction. The detector consists of flat tanks (60 cm by 90 cm and 10 cm thick) containing a calcium nitrate solution (20 percent Ca). Argon-37 is removed by purging the tanks with helium. The argon is purified and counted as already described. The efficiency of this detector and that of a similar tank filled with tetrachloroethylene has been measured with Pu-Be neutron source. The efficiencies were as follows: calcium nitrate tank  $3 \times 10^{-3}$ , and tetrachloroethylene tank  $2.4 \times 10^{-7}$  atoms Ar<sup>37</sup> per neutron. Measurements were performed with the neutron detectors in the room prior to building the 380,000 liter tank. The fast neutron background effect for the large tank was then estimated from its

surface area, the relative neutron efficiency for calcium nitrate and tetrachloroethylene, and the measured  $\text{Ar}^{37}$  production rate in the calcium nitrate detector. The  $\text{Ar}^{37}$  production rate derived in this manner for the solar neutrino detector is 0.1 atom per day. However, this fast neutron background effect can be greatly diminished by flooding the tank chamber with water to provide a fast neutron shield. The means of accomplishing this has already been described, see Figure 2.

There are background effects from internal natural alpha emitters. The most serious one arises from protons generated by the reaction  $\text{Cl}^{35}(\alpha, p)\text{Ar}^{38}$  that have sufficient energy to produce  $\text{Ar}^{37}$  by the  $\text{Cl}^{37}(p, n)\text{Ar}^{37}$  reaction. The yield of this reaction was measured by dissolving  $\text{Rn}^{222}$  in tetrachloroethylene, and removing the  $\text{Ar}^{37}$  by a helium purge. The yield measured was  $1.7 \times 10^{-10}$  atoms of  $\text{Ar}^{37}$  per  $\text{Rn}^{222}$  decay. The tetrachloroethylene was monitored for natural alpha emitters by sampling each railroad tank car at the time the tank was filled. This was accomplished by evaporating a liter sample and alpha-counting the residue. The tank walls (A-201-B steel) were carefully cleaned by shot blasting, and the piping was cleaned by acid dipping. The tank surface, and the piping was alpha-counted to determine the surface alpha emission rate. The total alpha emission rate from the metal surfaces and the liquid is approximately  $10^8$  alphas per day. Thus, the internal alpha production rate for  $\text{Ar}^{37}$  is estimated to be 0.02 atom per day from  $(\alpha, p)$  processes.

Internal alpha particles can also produce  $\text{Ar}^{37}$  by the direct reaction  $\text{S}^{34}(\alpha, n)\text{Ar}^{37}$  if the liquid contains sulfur. The yield for this reaction was measured by dissolving  $\text{Rn}^{222}$  in carbon disulfide, and removing the  $\text{Ar}^{37}$  by helium purge. A yield of  $1.8 \times 10^{-7}$   $\text{Ar}^{37}$  atoms per  $\text{Rn}^{222}$  decay was observed. The sulfur content of the tetrachloroethylene used is below 1 ppm, hence the



background effect from ( $\alpha, n$ ) reaction with sulfur is negligibly small, approximately  $10^{-5}$  Ar<sup>37</sup> atom per day.

It has been concluded from this examination of background processes that the sensitivity of the experiment can be improved by a factor of at least five without being limited by known background processes. The most serious restriction is from cosmic rays, and work is in progress to better evaluate the magnitude of this background.

There are several possible means of improving the over-all sensitivity of Ar<sup>37</sup> detection. These may be listed as follows.

1. Lower counter backgrounds by performing the measurements underground.  
An appreciable part of the counter background could arise from gamma radiation from the environment, and from cosmic ray interactions. If it is shown that the background arises from internal beta contamination, it will be necessary to find pure materials or use more careful cleaning techniques during counter assembly.
2. Use smaller counters with a correspondingly lower background counting rate. The size of the counter is now limited by the volume of the argon sample that is counted ( $0.5 \text{ cm}^3$ ). Most of this volume is air argon that arises either from inleakage of air, or from sources within the tank. Sources of this air argon are being actively sought. However, in the event that this source cannot be eliminated then one could consider argon isotope separation to remove the Ar<sup>40</sup>. The isotopes of argon could be separated by gas phase chromatography using a sufficiently long charcoal column.
3. Employ various techniques to distinguish the Ar<sup>37</sup> decays from background processes that may arise from the walls of the counter. The possibilities are: (a) use an internal anticoincidence counter; (b) provide a grid

near the wall biased to eliminate ions arising from the wall; or

(c) identify pulses initiated in the gas by pulse shape.

4. Perform a series of experimental runs and sum the pulse height spectra. The resulting spectrum would allow a more sensitive search for a peak at 2.8 keV from Ar<sup>37</sup> decay. This approach would be very effective if the counter background were essentially zero, since it would constitute a nearly continuous observation of the solar neutrino flux.

These various approaches are being examined. The obvious approach would be to build a larger detector, but this is not being seriously considered at present.

#### Other Radiochemical Methods for Detecting Neutrinos

The results of the Cl<sup>37</sup> experiment make it appear very likely that the flux of energetic neutrinos from B<sup>8</sup> decay in the sun is less than  $10^6 \text{ cm}^{-2} \text{ sec}^{-1}$ . One should therefore develop another neutrino detection technique capable of observing low energy neutrinos. The radiochemical method, with its high sensitivity, appears to be a fruitful approach to explore. Though it is also possible to conceive of detectors based upon the scattering of neutrinos by electrons, as Professor Reines has suggested at this conference.

Dr. Keither Rowley and the author have made search for possible neutrino capture reactions that would be suitable for a radiochemical detection system. A list was compiled of all radioactive isotopes produced from stable isotopes by ( $\gamma, e^-$ ) reaction with a threshold below 1 MeV. Out of this list we chose only those cases in which the transition was allowed ( $\log ft < 6$ ), and resulted in a radioactive product with a half-life under 5 years but more than 20 minutes. An additional chemical restriction was imposed. It seems unlikely that one will be able to separate a few atoms of

a rare earth element from many tons of a neighboring rare earth element, so these cases were eliminated from further consideration. After these restrictions were imposed, the number of suitable ( $\nu, e^-$ ) reactions was extremely limited, and with one exception, were the ones already discussed in the literature. The reactions selected are summarized in Table IV. In this table is listed the calculated neutrino capture rate from Dr. Bahcall's paper, and the number of tons of the element required for one capture per day.

One must then select from this short list of possibilities, the most suitable reaction. The major considerations are, the availability of the target element, and the sensitivity of counting the radioactive product. One has confidence that an efficient chemical separation technique can be developed.

The  $Rb^{87}$  reaction, that was suggested by Goldhaber,<sup>16</sup> takes advantage of the isomeric state in  $Sr^{87}$ . This reaction has the lowest threshold of those listed. The chemical separation of strontium from an aqueous solution of a rubidium salt could be readily accomplished with a chelating resin. A rapid chemical separation is required, since the half-life of  $Sr^{87m}$  is only 2.8 hours. In fact a chemical processing system would have to be used that would remove a sample every three hours or so. The counting of the 388 keV gamma (or its conversion electron) from  $Sr^{87m}$  decay could be accomplished with a silicon or germanium detector to achieve high resolution. Rubidium is a rare and expensive element, but ton quantities may be obtained.

The very attractive reaction  $Ga^{71}(\nu, e^-)Ge^{71}$  has been discussed by several authors,<sup>17</sup> and by Professor Zatsepin at this conference. The main difficulty again is the very high cost and availability of ton quantities of gallium. The chemical separation can certainly be accomplished in an aqueous solution.

The  $\text{Li}^7(\nu, e^-)\text{Be}^7$  reaction is an attractive possibility. Since the transition is superallowed, and lithium is a light element, the quantity required is rather modest. However, counting  $\text{Be}^7$  radioactivity with high sensitivity is rather difficult. There are several techniques that could be applied: (1) count the very low energy Auger electrons (280 eV); (2) count the 480 keV gamma ray that occurs in 11 percent of the decays; or (3) count the recoiling lithium ion (57 eV).<sup>18</sup> The carrier-free chemical separation of beryllium from an aqueous solution of a lithium salt could be accomplished by using a chelating resin, or by the use of a ferric hydroxide scavenger. Hence, if a counting system could be devised to observe a count per day of  $\text{Be}^7$  a large scale lithium solar neutrino detector appears to be entirely feasible. Dr. Bahcall has pointed out in this conference that a measurement of the solar neutrino capture rate in  $\text{Li}^7$  would be especially valuable for understanding the processes in the sun, particularly if the solar neutrino capture rate in  $\text{Cl}^{37}$  were observed.

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Table I

Calculated Neutrino Capture Rate in Chlorine-37 from Various Neutrino Sources in the Sun

Source	Flux at Earth, <sup>a</sup> $\phi$ (cm <sup>-2</sup> sec <sup>-1</sup> )	Cross Section, <sup>b</sup> $\sigma$ (cm <sup>2</sup> )	Capture Rate in Chlorine-37 (sec <sup>-1</sup> · 10 <sup>36</sup> )
H + H → D + e <sup>+</sup> + $\nu$	6.35 × 10 <sup>10</sup>	0	0.0
H + H → D + $\nu$	1.65 × 10 <sup>8</sup>	1.72 × 10 <sup>-45</sup>	0.28
Be <sup>7</sup> decay	2.9 × 10 <sup>9</sup>	2.9 × 10 <sup>-46</sup>	0.84
B <sup>8</sup> decay	3.6 × 10 <sup>6</sup>	1.35 × 10 <sup>-42</sup>	4.9
N <sup>13</sup> decay	2.2 × 10 <sup>8</sup>	2.1 × 10 <sup>-46</sup>	0.05
O <sup>15</sup> decay	2.2 × 10 <sup>8</sup>	7.8 × 10 <sup>-46</sup>	0.17

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$$\sum \phi \sigma = 6.2 \times 10^{-36} \text{ sec}^{-1}$$

<sup>a</sup> Fluxes from paper presented at this conference by J. N. Bahcall.

<sup>b</sup> From Bahcall.<sup>5</sup>

Table II  
 Recovery of Ar<sup>37</sup> Produced by Neutron Irradiation

Volume of Helium Passed, liters	Ar <sup>36</sup> Recovery, percent	Ar <sup>37</sup> Recovered dis. day <sup>-1</sup>
350,000	90.6	63.4 ± 3.6
260,000	6.2	2.3 ± 1.1
340,000	0.7	0.7 ± 0.5
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950,000	97.5	66.4 ± 3.8 <sup>‡</sup>

<sup>‡</sup> Total Ar<sup>37</sup> production from Ra-Be neutron source =  $(7.5 \pm 0.4) \times 10^{-7}$   
 Ar<sup>37</sup> atoms per neutron.



Table III

## Limits on the Extraterrestrial Neutrino Flux

Energy MeV	Cross Section, <sup>5,10</sup> cm <sup>2</sup>	Flux Limit cm <sup>-2</sup> sec <sup>-1</sup>	Energy Density ØE/c, keV cm <sup>-3</sup>
1	0.55 x 10 <sup>-45</sup>	5 x 10 <sup>9</sup>	180
5	1.5 x 10 <sup>-43</sup>	2 x 10 <sup>7</sup>	3
10	2.3 x 10 <sup>-42</sup>	13 x 10 <sup>5</sup>	0.4
100	~ 7 x 10 <sup>-39</sup>	4 x 10 <sup>2</sup>	0.013

Table IV

## Neutrino Capture Reactions

Reaction	Half-life	Threshold, keV	$\sum \sigma \times 10^{36} \text{ sec}^{-1}$	Tons of the Element Required for 1 event/day
$\text{Rb}^{87}(\nu, e^-)\text{Sr}^{87\text{m}}$	2.8 hours	115	490	12
$\text{Mn}^{55}(\nu, e^-)\text{Fe}^{55}$	2.6 years	231	3.8	280
$\text{Ga}^{71}(\nu, e^-)\text{Ge}^{71}$	11 days	233	290	11
$\text{V}^{51}(\nu, e^-)\text{Cr}^{51}$	28 days	752	3.0	330
$\text{Cl}^{37}(\nu, e^-)\text{Ar}^{37}$	35 days	816	6.2	430
$\text{Li}^7(\nu, e^-)\text{Be}^7$	53 days	862	36	4

### Figure Captions

- Figure 1. Schematic arrangement of the apparatus.
- Figure 2. The arrangement of the Brookhaven solar neutrino detector in the Homestake Gold Mine, Lead, South Dakota, U.S.A.
- Figure 3. Pulse height spectra from run no. 9.
- Figure 4. Pulse height spectra for run no. 13.
- Figure 5. Chronological occurrence of the counts within the Ar<sup>37</sup> peak position in run no. 13.









