SEARCH FOR NEUTRINOS FROM THE SUN*

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(Received 16 April 1968)

A search was made for solar neutrinos with a detector based upon the reaction $Cl^{37}(\nu, e^-)Ar^{37}$. The upper limit of the product of the neutrino flux and the cross sections for all sources of neutrinos was 3×10^{-36} sec^{-1} per Cl^{37} atom. It was concluded specifically that the flux of neutrinos from B^8 decay in the sun was equal to or less than 2×10^6 cm^{-2} sec^{-1} at the earth, and that less than 9% of the sun's energy is produced by the carbon-nitrogen cycle.

Recent solar-model calculations have indicated that the sun is emitting a measurable flux of neutrinos from decay of B^8 in the interior.[1-8] The possibility of observing these energetic neutrinos has stimulated the construction of four separate neutrino detectors.[9] This paper will present the results of initial measurements with a detection system based upon the neutrino capture reaction $Cl^{37}(\nu, e^-)Ar^{37}$. It was pointed out by Bahcall[10] that the energetic neutrinos from B^8 would feed the analog state of Ar^{37} (a superallowed transition) that lies 5.15 MeV above the ground state. The importance of the contribution of the B^8 neutrino flux is readily seen from the neutrino-capture cross sections and the solar neutrino fluxes given in Table I. The tabulated fluxes were taken from the calculations of Bahcall and Shaviv,[8] who studied the effect of errors in the parameters—solar composition, luminosity, opacity, and nuclear reaction cross sections. These authors have placed a probable error of 60% on the calculated B^8 flux. Their predicted B^8 flux for mean values of the various parameters agrees well with the independent calculations of Ezer and Cameron.[5] On the basis of these predictions, the total solar-neutrino-capture rate in 520 metric tons of chlorine would be in the range of 2 to 7 per day.

The detector design.—A detection system that contains 390 000 liters (520 tons chlorine) of liquid tetrachloroethylene, C_2Cl_4, in a horizontal cylindrical tank was built along the lines proposed earlier.[11] The system is located 4850 ft underground [4400 m (w.e.)] in the Homestake gold mine at Lead, South Dakota. It is essential to place the detector underground to reduce the production of Ar^{37} from (p,n) reactions by protons formed in cosmic-ray muon interactions. The rate of Ar^{37} production in the liquid by cosmic-ray muons at this location is estimated to be 0.1 Ar^{37} atom per day.[11] Background effects from internal α contaminations and fast neutrons from the surrounding rock wall are low. The total Ar^{37} production from all background processes is less than 0.2 Ar^{37} atom per day, which is well below the rate expected from solar neutrinos.

Neutrino detection depends upon removing the Ar^{37} from a large volume of liquid contained in a sealed tank, and observing the decay of Ar^{37} (35-day half-life) in a small proportional counter (0.5 cm^3). It is therefore necessary to have an efficient method of removing a fraction of a cubic centimeter of argon from 390 000 liters of C_2Cl_4. The Ar^{37} activity is removed by purging with helium gas. Liquid is pumped uniformly

Table I. Solar neutrino fluxes and cross sections for the reaction $Cl^{37}(\nu, e^-)Ar^{37}$.

Neutrino source	Cross section[a,b] (cm^2)	Neutrino flux[c] at the earth (cm^{-2} sec^{-1})	$10^{35}\sigma\varphi$ (sec^{-1})
$H + H + e^- \rightarrow D + \nu$	1.72×10^{-45}	1.7×10^8	0.03
Be^7 decay	2.9×10^{-46}	3.9×10^9	0.11
B^8 decay	1.35×10^{-42}	$1.3(1 \pm 0.6) \times 10^7$	$1.8(1 \pm 0.6)$
N^{13} decay	2.1×10^{-46}	1.0×10^9	0.02
O^{15} decay	7.8×10^{-46}	1.0×10^9	0.08
			$\sum \varphi\sigma = 2.0(1 \pm 0.6) \times 10^{-35}$ sec^{-1}

[a]Ref. 4.   [b]Ref. 10.   [c]Ref. 8.

from the bottom of the tank and returned to the
tank through a series of 40 eductors arranged
along two horizontal header pipes inside the tank.
The eductors aspirate the helium from the gas
space (2000 liters) above the liquid, and mix it
as small bubbles with the liquid in the tank. The
pump and eductor system passes helium through
the liquid at a total rate of 9000 liters per min-
ute maintaining an effective equilibrium between
the argon dissolved in the liquid and the argon in
the gas phase.

Argon is extracted by circulating the helium
from the tank through an argon extraction sys-
tem. Gas flow is again achieved by a pair of
eductors in the tank system, and they maintain a
flow rate of 310 liters per minute through the ar-
gon extraction system. The tetrachloroethylene
vapor is removed by a condenser at $-40°C$ fol-
lowed by a bed of molecular sieve adsorber at
room temperature. The helium then passes
through a charcoal bed at $77°K$ to adsorb the ar-
gon, and is finally returned to the tank. This ar-
rangement is shown schematically in Fig. 1.
The apparatus is located in three separate rooms

in the mine as indicated in the diagram.

The argon sample adsorbed on the charcoal
trap is removed by warming the charcoal while
a current of helium is passed through it. The ar-
gon and other rare gases from the effluent gas
stream are collected on a small liquid-nitrogen-
cooled charcoal trap (1 cm diam by 10 cm long).
Finally, the gases from this trap are desorbed
and heated over titanium metal at $1000°C$ to re-
move all traces of chemically reactive gases.
The resulting rare gas contains krypton and xe-
non in addition to argon. These higher rare gas-
es were dissolved from the atmosphere during
exposure of the liquid during the various manu-
facturing, storage, and transfer operations.
Krypton and xenon are much more soluble in tet-
rachloroethylene than argon, and, therefore,
they are more slowly removed from the liquid
by sweeping with helium. Since the volume of
krypton and xenon in an experimental run is com-
parable with or exceeds the volume of argon, it
is necessary to remove these higher rare gases
from the sample. A more important considera-
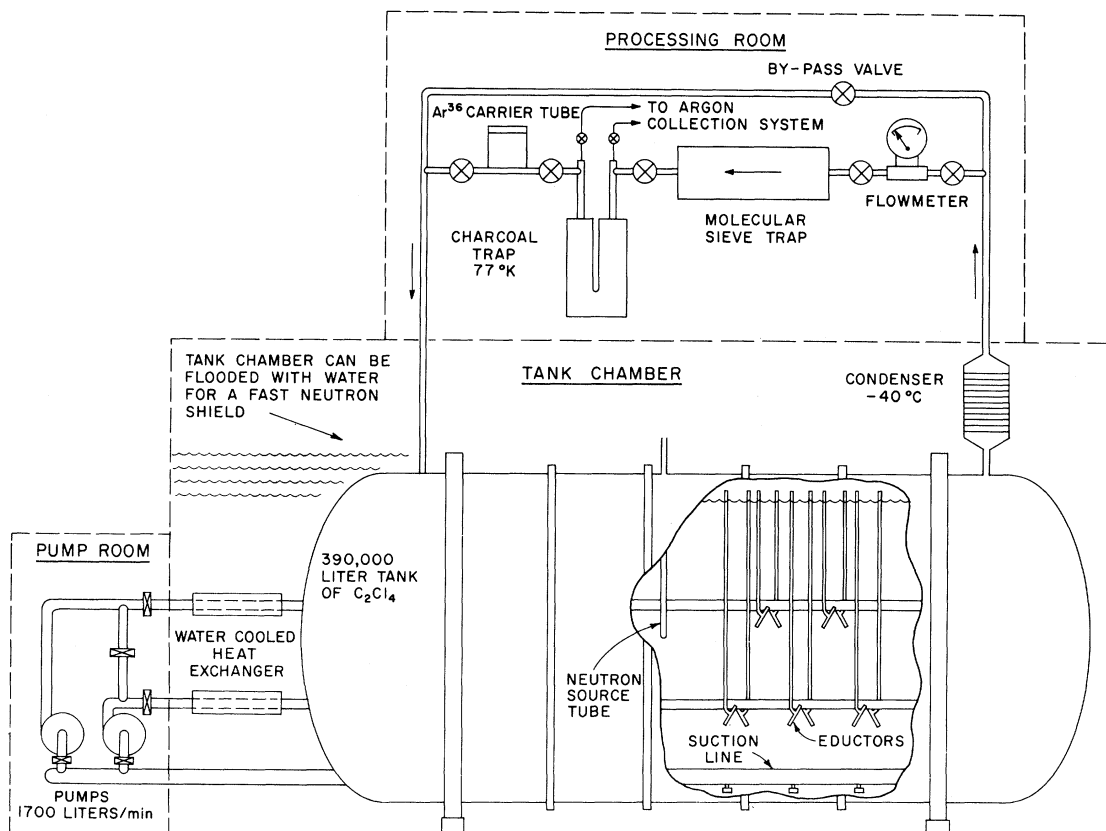tion is that atmospheric krypton contains the

FIG. 1. Schematic arrangement of the Brookhaven solar neutrino dector.

10.8-yr fission product Kr^{85}. The rare gases recovered from the tank are therefore separated by gas chromatography. To insure complete removal of krypton from the argon sample, a second gas chromatographic separation is made of the argon fraction. Experience has shown that these two successive chromatographic separations reduce krypton concentration in the argon sample to less than 10^{-8} parts per volume. The entire purified argon sample is counted in a small proportional counter that will be described later.

Argon recovery tests.—After the air and air argon had been removed from the system by prolonged sweeping with helium, the argon recovery efficiency of the system was measured by an isotope dilution method. A measured volume of 99.9% Ar^{36} was introduced into the tank and dissolved in the liquid with the eductor system. It was then recovered by six separate purging operations. The Ar^{36} recovered from each purge was determined by a volumetric and argon mass-ratio measurement. It was found that the volume of Ar^{36} in the tank dropped exponentially with the volume of helium circulated according to

$$v(Ar)/v_0(Ar) = e^{-7.21 \times 10^{-6} V(He)}.$$

where $v_0(Ar)$ is the initial volume of Ar^{36} and $v(Ar)$ is the volume remaining after $V(He)$ liters of helium have passed through the extraction system. This test showed that a 95% recovery of argon from the tank can be achieved by circulating 0.42 million liters of helium through the extraction system, which requires a period of 22 h.

Another test of the argon recovery from the tank was performed with Ar^{37} activity produced in the tank by a fast-neutron irradiation. A Ra-Be neutron source with a total neutron emission rate of 7.38×10^4 neutrons sec^{-1} was inserted in a re-entrant iron pipe that reaches to the center of the tank. The liquid was irradiated with this source for 0.703 days producing Ar^{37} in the liquid by the reaction $Cl^{37}(p,n)Ar^{37}$ from the protons produced in the liquid principally by the reaction $Cl^{35}(n,p)S^{35}$. Carrier Ar^{36} was introduced (1.18 std cc) and the tank was swept three successive times with helium in which the volumes passed were, respectively, 0.35, 0.26, and 0.34 millions of liters of helium. The recovered argon was purified and counted following the procedures given below. The Ar^{37} activities in the three separate purges were found to be 63.4 ± 3.6, 2.3 ± 1.1, and 0.7 ± 0.5 disintegrations per

day at the end of the neutron irradiation. The total Ar^{37} production rate observed in this experiment was $(7.5 \pm 0.4) \times 10^{-7}$ Ar^{37} atom per neutron. This production rate compared favorably with similar measurements in containers of smaller diameters (29 and 120 cm) which gave yields of 3.0×10^{-7} and 6.4×10^{-7} Ar^{37} atom per neutron, respectively. The Ar^{36} recoveries from each of the three successive purges were 90.6, 6.2, and 0.7 %, matching closely the Ar^{37} recoveries.

One might question whether Ar^{37} produced by the (ν, e^-) reaction would also be removed efficiently, since it would initially have a lower recoil energy than Ar^{37} produced by the (p, n) reaction. The Ar^{37} recoil energy resulting from neutrino capture ranges from 11 to over 1000 eV for neutrino energies of 1 to 10 MeV. These recoil energies are sufficient to assure that the Ar^{37} ion formed would be free of the parent molecule, and, therefore, it would be expected to behave chemically similarly to an Ar^{37} atom produced by the (p, n) reaction. Once an Ar^{37} atom exists as a free atom it will mix with the carrier Ar^{36} present in the liquid (10^{10} atoms cm^{-3}) and be removed by the helium purge.

Counting.—The argon sample is counted in a small proportional counter with an active volume 3 cm long and 0.5 cm in diameter. A small amount of methane is added to the argon to improve the counting characteristics of the gas. The counter cathode was constructed of zone-refined iron and the exterior envelope is made of silica glass. A thin window in the envelope is located at the end of the counter to facilitate energy calibration of the counter with Fe^{55} x rays. The counter is shielded from external radiations by a cylindrical iron shield 30 cm thick lined with a ring of 5-cm-diam proportional counters for registering cosmic-ray muons. The argon counter is held in the well of a 12.5- by 12.5-cm sodium-iodide scintillation counter located inside the ring counters. Events in anticoincidence with both the ring counters and the scintillation counter are recorded on a 100-channel pulse-height analyzer.[12] The pulse-height and time distribution of the events are recorded on paper tape. Each anticoincidence pulse is displayed on a storage oscilloscope and photographed to allow examination of each pulse shape to insure that it has a proper shape and is not caused by electrical noise. The counter had a 28% resolution (full width at half-maximum) for the 2.8-keV Auger electrons from the Ar^{37} decay. The operating voltage and amplifier gain are adjust-
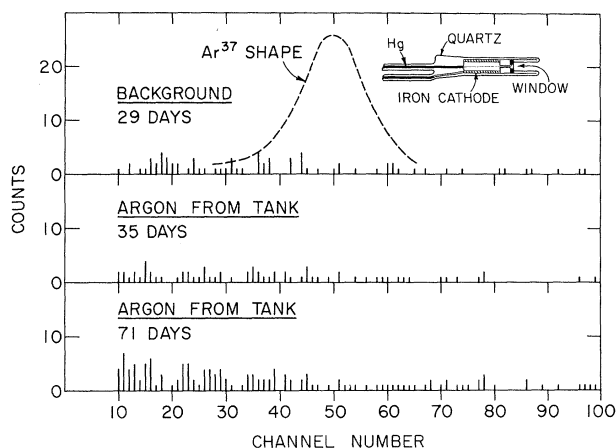
FIG. 2. Pulse-height spectra.

ed to place the center of the 2.8-keV Ar^{37} peak at channel 50 in the spectrum. The background counting rate in the 14 channels centered around channel 50 is 0.3 count per day (see Fig. 2). The efficiency of the counter was determined by filling with argon containing a known amount of Ar^{37}. Its efficiency for Ar^{37} is 51 % for the 14 channels centered about channel 50.

Results and discussion. – Two experimental runs have been performed. In both experiments a measured volume of Ar^{36} was introduced into the tank at the start of the period of exposure, and mixed into the liquid for a period of approximately two hours with the eductor system. During the period of exposure the pumps were not operated. A positive pressure of helium of approximately 250 mm of Hg exists in the tank at all times.

The first exposure was 48 days. The tank was purged with 0.50 million liters of helium. A volume of 1.27 std cc of argon was recovered from the tank, and this volume contained 94 % of the carrier Ar^{36} introduced at the start of the exposure. It was counted for 39 days and the total number of counts observed in the Ar^{37} peak position (full width at half-maximum) in the pulse-height spectrum was 22 counts. This rate is to be compared with a background rate of 31 ± 10 counts for this period. The neutrino-capture rate in the tank deduced from the exposure, counter efficiency, and argon recovery from this experiment was (−1.1 ± 1.4) per day.

A second exposure was made for 110 days from 23 June to 11 October 1967. The tank was purged with 0.53 million liters of helium yielding 0.62 cm^3 of argon with a 95% recovery of the added carrier Ar^{36}. The pulse-height spectra are shown in Fig. 2 for the first 35 days of counting

and also for a total period of 71 days. This rate can be compared with the background rate for the counter filled with Ar^{36} purified in an identical manner (shown in Fig. 2). It may be seen from the pulse-height spectrum for the first 35 days of counting that 11 ± 3 counts were observed in the 14 channels where Ar^{37} should appear. The counter background for this period of time corresponded to 12 ± 4 counts. Thus, there is no increase in counts from the sample over that expected from background counting rate of the counter. One would deduce from these rates that the neutrino-capture rate in 610 tons of tetrachloroethylene was equal to or less than 0.5 per day based upon one standard deviation. A similar limit can be obtained if one examines the shape of the pulse-height spectrum for extra counts in the 14 channels centered about channel 50 in the first 35-day count.

This limit, expressed as

$$\sum \varphi \sigma \leq 0.3 \times 10^{-35} \ \text{sec}^{-1} \ \text{per Cl}^{37} \ \text{atom,}$$

can be compared with the predicted value of (2.0 ± 1.2) × 10^{-35} sec^{-1} per Cl^{37} atom (Table I). It may be seen that this limit is approximately a factor of 7 below that expected from these solar-model calculations. From this limit and the cross section for B^8 neutrinos given in Table I, it may be concluded that the flux of B^8 neutrinos at the earth is equal to or less than 2 × 10^6 cm^{-2} sec^{-1}. It may be pointed out that if one accepted all of the 11 counts in the spectrum for the 35-day count as real events, making no allowance for background, then the flux–cross-section product limit would be 0.6 × 10^{-35} sec^{-1} per Cl^{37} atom.

The solar-model calculation of the flux of B^8 neutrinos is dependent upon the nuclear cross sections, solar composition, solar age and luminosity, and the opacity of solar material. The effect of each of these parameters has been studied, and the present results show that the solar B^8 neutrino flux is outside the present error limits if the uncertainties are treated as probable errors.[6-8] In the following article[13] Bahcall, Bahcall, and Shaviv have re-evaluated the solar neutrino fluxes taking into account a new value for the heavy element composition of the sun, and a new rate for the reaction H(H, $e^+\nu$)D.

Since this experiment is the first one with sufficient sensitivity to detect solar neutrinos from the carbon-nitrogen cycle, it is interesting to draw a conclusion about this energy cycle. Bahcall[4] has calculated the total flux–cross-section

product for the carbon-nitrogen cycle to be 3.5 $\times 10^{-35}$ sec^{-1} per Cl^{37} atom, based on this cycle being the only source of the sun's energy. With the limit given above one can conclude that less than 9% of the sun's energy is produced by the carbon-nitrogen cycle.

It is possible to improve the sensitivity of the present experiment by reducing the background of the counter. However, background effects from cosmic-ray muons will eventually limit the detection sensitivity of the experiment at its present location. Detailed studies of the cosmic-ray background are in progress.

The authors would like to thank Professor W. A. Fowler and Professor John N. Bahcall for their initial and continual encouragement in planning this experiment. We would like to acknowledge Professor A. G. W. Cameron's constant interest extending over many years. We are indebted to the Homestake Mining Company for allowing us to build the experiment in their mine, and for their generous assistance in solving many technical problems in the construction of the apparatus. We would like to acknowledge the many useful suggestions and direct assistance from the members of the staff of Brookhaven National Laboratory.

*Research performed under the auspices of the U. S. Atomic Energy Commission.

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PRESENT STATUS OF THE THEORETICAL PREDICTIONS FOR THE ^{36}Cl SOLAR-NEUTRINO EXPERIMENT*

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(Received 8 April 1968)

The theoretical predictions for the ^{37}Cl solar-neutrino experiment are summarized and compared with the experimental results of Davis, Harmer, and Hoffman. Three important conclusions about the sun are shown to follow.

The experiment of Davis, Harmer, and Hoffman,[1,2] designed to detect solar neutrinos with a ^{37}Cl target, has prompted a continuing investigation[3-7] of the accuracy with which the flux of neutrinos produced by nuclear reactions in the sun's interior can be predicted. We report here calculations of the solar-neutrino fluxes made using the more accurate rate for the proton-proton reaction recently derived by Bahcall and May[8] and the improved determination of the abundance ra-

tio of heavy elements to hydrogen recently obtained by Lambert and Warner.[9] We also discuss some of the important, recognized uncertainties that influence the predictions of the solar-neutrino fluxes and conclude that the present results of Davis, Harmer, and Hoffman[1] are not in obvious conflict with the theory of stellar structure. We show, however, that a counting rate of less than $0.03 \times 10^{-35}/^{37}$Cl atom sec would cast serious doubt on the correctness of current ideas con-