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Attempt to Detect the Antineutrinos from a Nuclear Reactor by the $Cl^{37}(\bar{\nu},e^-)A^{37}$ Reaction*

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Tanks containing 200 and 3900 liters of carbon tetrachloride were irradiated outside of the shield of the Brookhaven reactor in an attempt to induce the reaction $Cl^{37}(\bar{\nu},e^-)A^{37}$ with fission product antineutrinos. The experiments serve to place an upper limit on the antineutrino capture cross section for the reaction of 2×10^{-42} cm² per atom. Cosmic-ray-induced A^{37} was observed and the production rate measured at 14 100 feet altitude and sea level. Measurements with the 3900-liter container shielded from cosmic rays with 19 feet of earth permit placing an upper limit on the neutrino flux from the sun.

I. INTRODUCTION

THERE have been a number of experiments performed in the past to detect the neutrino by scattering processes and nuclear interactions.[1] The most sensitive of these experiments serve to place a limit on the scattering cross section for neutrinos on electrons of less than 14×10^{-40} cm²/electron and for nuclear interaction of less than 10^{-30} cm²/atom. Recently Reines and Cowan of the Los Alamos Laboratory performed an experiment with a large hydrocarbon liquid scintillator having a high sensitivity for detecting the interaction $p(\bar{\nu},e^+)n$ within the liquid.[2] Measurements were made with this scintillator located adjacent to the Hanford reactor within a shield designed to absorb other radiations from the reactor to which the scintillator was sensitive. Under these conditions they observed an increase in counting rate of the scintillator of 0.41 ± 0.20 delayed count per minute when the reactor was operating over that observed with the reactor off. This increase in counting rate, if ascribed to the process $p(\bar{\nu},e^+)n$, corresponds to a cross section of $(12\pm6)\times10^{-44}$ cm²/atom.

In 1946 Pontecorvo[3] suggested a radiochemical method of detecting the neutrino by employing the reaction $Cl^{37}(\bar{\nu},e^-)A^{37}$. The experiment involved irradiating a large volume of carbon tetrachloride near a nuclear reactor, removing the A^{37} by physical methods, and counting the electron capture decay of this isotope. Alvarez[4] considered the use of this method of detecting the neutrino in detail, and proposed an extended experiment capable of detecting the theoretically expected cross section for neutrinos of 2×10^{-45} cm²/atom.

The reaction $Cl^{37}(\nu,e^-)A^{37}$ is the inverse process to the 34-day electron capture decay of A^{37}. In this decay a neutrino (ν) is emitted which may be formally distinguished from an antineutrino ($\bar{\nu}$) which accompanies negative beta emission. A nuclear reactor emits antineutrinos which arise from the negative beta decays of fission products. In our experiment an attempt is made to observe an inverse electron capture process which requires neutrinos, using a source emitting antineutrinos. If neutrinos and antineutrinos are identical in their interactions with nucleons one should be able to observe the process upon carrying the experiment to the required sensitivity. However, if neutrinos and antineutrinos differ in their interactions with nucleons one would not expect to induce the reaction $Cl^{37}(\bar{\nu},e^-)A^{37}$. A positive experiment of this type would show that these particles are not to be distinguished in their nuclear reactions. A negative experiment carried to the required sensitivity would indicate that neutrinos and antineutrinos differ in their nuclear reactions, or that the present theory of beta decay is incorrect. The present theory of beta decay and the principle of detailed balancing lead to a reliable calculated cross section for the inverse process. The only evidence concerning the nuclear interactions of neutrinos and antineutrinos comes at present solely from a study of the half-life of the double beta-decay process. These studies have indicated that neutrinos and antineutrinos do differ in their interactions with nucleons.[5]

An experiment has been performed in which a 200-liter (55-gallon) drum of carbon tetrachloride was irradiated outside the shield of the Brookhaven reactor. The argon was removed by sweeping with helium gas and counted. Further measurements were carried out with a 3900-liter (1000-gallon) tank of carbon tetrachloride. As a result of these measurements one can place an upper limit on the cross section for the reaction of 2×10^{-42} cm²/atom for fission product antineutrinos. A background of A^{37} activity was produced in the tank by cosmic-ray interactions and in the present experiment prevented observing lower values of the cross section. The purpose of this paper is to give the results of these measurements.

The present experiment was not sensitive enough to

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[1] The experiments are reviewed by H. R. Crane, Revs. Modern Phys. **20**, 278 (1948); J. H. Barrett, Phys. Rev. **79**. 907 (1950); Cowan, Reines, and Harrison, Phys. Rev. **96**, 1294 (1954).

[2] F. Reines and C. L. Cowan, Jr., Phys. Rev. **92**, 830 (1953).

[3] B. Pontecorvo, Chalk River Laboratory Report PD-205, 1948 (unpublished).

[4] L. W. Alvarez, University of California Radiation Laboratory Report UCRL 328, 1949 (unpublished).

[5] Reviewed by E. J. Konopinski and L. M. Langer, Ann. Rev. Nuc. Sci. **2**, 261 (1953).

detect the theoretically computed cross section and therefore conclusions could not be drawn concerning the correctness of beta decay theory, or the identity of neutrinos and antineutrinos. The present experiment will be continued in an effort to accomplish this purpose.

II. EXPERIMENTAL

Procedure for Removing Argon-37

Argon-37 produced by nuclear reactions in the carbon tetrachloride can be removed from the liquid by sweeping it with helium gas. The argon can then be removed from the helium by passing the gas through a charcoal trap cooled with liquid nitrogen ($-196°C$) which adsorbs the argon quantitatively and allows the helium to pass. The carbon tetrachloride was contained in a 200-liter drum and 3900-liter tank. Each was equipped with a stirrer and a valved tube for introducing the helium into the bottom of the container and an exit valve and tube at the top. Helium gas from a cylinder was purified by passing it through a charcoal trap cooled with liquid nitrogen and then passed into the container. Upon leaving the container the gas was passed through condensation traps to remove carbon tetrachloride vapor. Following these the gas passed through two charcoal traps in series. The first was cooled with solid carbon dioxide ($-78°C$) and served to remove radon and xenon gas which might arise from contaminants in the system. The second charcoal trap was cooled with liquid nitrogen and retained the argon and krypton. Krypton was partially removed by the first charcoal trap at the flow rates and quantities of helium gas used in the sweeping. After completion of the sweeping the second charcoal trap was evacuated to remove the occluded helium, and closed off. The argon contained in this charcoal trap contained some radioactive rare gases that had to be removed before installing the gas in a Geiger-Müller counter. The ones that have been found present are radon, arising from a small amount of radium impurity in the charcoal, and fission product krypton, particularly 9.5-year Kr^{85}, which could be introduced into the connecting tubes on the tanks during the irradiation near the reactor. These were removed by sweeping the gas isolated above with helium through another charcoal trap cooled with solid carbon dioxide into a small charcoal trap cooled with liquid nitrogen. The gas contained in the small trap was evacuated cold to remove the occluded helium, and then desorbed by heating to 200°C. The argon gas was removed from the charcoal trap, heated over Ti metal to 900°C, and installed along with quench gas as the counting gas in a Geiger-Müller counter.

Small amounts of argon could be removed from a 200-liter drum of carbon tetrachloride by sweeping it with approximately 400 liters of helium at a rate of 1 to 2 liters per minute. The 3900-liter tank could be swept out with approximately 4000 liters of helium at a rate of 5 to 7 liters per minute. The efficiency of the sweeping operation was tested in two ways. First, small measured amounts of argon gas (20 to 0.2 cm^3) were introduced into the container, allowed to remain for several days and then removed by the sweeping procedure and the amount recovered was measured. A second method takes advantage of the fact that A^{37} is produced in carbon tetrachloride by cosmic rays. This activity was removed by the sweeping procedure described, and counted. Immediately afterward a second identical sweeping operation was conducted which yielded less than ten percent of the A^{37} activity obtained in the first sweeping. It was also demonstrated that the charcoal trap removes the argon quantitatively. This was shown by sweeping a 320-liter container in which A^{37} was produced in the carbon tetrachloride liquid by irradiation with fast neutrons. Here A^{37} was produced by the reaction $Cl^{37}(p,n)A^{37}$ with protons produced by (n,p) reactions on Cl^{35}. A sweeping operation was carried out by using two charcoal traps in series. The first trap contained 120 counts per minute of A^{37} and the second contained less than 0.1 count per minute.

It was concluded from these experiments that the sweeping procedures used will remove over ninety percent of the A^{37} activity free from other rare-gas radioactive contaminants.

Irradiation at the Brookhaven Reactor

The Brookhaven reactor operates at a power level 20–25 megawatts producing approximately 4×10^{18} antineutrinos per second. The graphite cube is 25 feet on a side and is surrounded by a concrete and iron shield five feet thick. The containers of carbon tetrachloride were placed outside of the shield and irradiated for 36 to 75 days. The 200-liter drum and the 3900-liter tank were located 29 feet from the center of the reactor and received a flux of 3 to 4×10^{11} antineutrinos per cm^2 per second. The 200-liter drum was surrounded by a shield of iron loaded concrete eight inches thick. In addition the drum was shielded from above by two extensive balconies with a combined thickness of 16 inches of concrete. The 3900-liter tank was not shielded. However, both the tank and the drum were effectively shielded on one side by their proximity to the large reactor shield.

Counters

Small Geiger-Müller counters made of stainless steel and cold-rolled steel were used. These were 0.48 cm in diameter and 12 cm long and contained a 0.050-mm tungsten center wire. They were filled with 200 to 400 mm of argon pressure and 25 mm of ethyl acetate quench gas. These counters operated around 1000 volts and had plateaus 100 volts long with a slope of less than five percent per 100 volts. The counter was surrounded by a ring of six anticoincidence counters

TABLE I. Summary of results.

	200-liter container			3900-liter container	
Location during irradiation	Adjacent to reactor	Laboratory	Mt. Evans	Adjacent to reactor	Buried 19 feet underground
Amount of shielding over container, g/cm^2	120	None	None	None	970
Duration of irradiation, days	75	77	62	36	41
Average power level of reactor, in megawatts	17	...	...	21	...
Average antineutrino flux/cm^2/sec	3.4×10^{11}	...	...	4.4×10^{11}	...
Saturation counting rate, disintegration/min	<0.03	<0.03	3.5 ± 0.5	1.23 ± 0.06	<0.05

2.5 cm in diameter and 30 cm long. The counting was carried out in an iron shield seven inches thick. The background counting rate of the counters may be summarized as follows: outside of shield 15 counts per minute, inside of shield 4.3 counts per minute, and in anticoincidence 0.15 to 0.25 count per minute. A given counter was found to give a reproducible background counting rate upon changing the filling gas. An increase in counting rate of ten percent of the background counting could be detected.

III. RESULTS AND DISCUSSION

Two measurements were made with the 200-liter container of carbon tetrachloride. In one the drum was placed close to the reactor, and in the second it was placed in the laboratory 120 feet from the reactor. The A^{37} activity in both of these drums was below the amount detectable with the counters used (see Table I). From the result an upper limit of 2×10^{-42} cm^2/atom can be set for the cross section for the reaction $Cl^{37}(\bar{\nu},e^-)A^{37}$ for fission product antineutrinos. The antineutrino flux from the reactor was computed from the average power level of the reactor and the result of Way and Wigner[6] who showed that on the average 6.1 beta decays follow each fission event. The experiments with these containers showed that it was possible to remove quantitatively small amounts of A^{37} from large quantities of carbon tetrachloride by sweeping with helium and that background effects producing A^{37} activity are below the sensitivity of these experiments. It then appeared possible to continue the experiments with increased sensitivity.

The most serious background effect expected in this experiment arises from the production of A^{37} by energetic protons by the reaction $Cl^{37}(p,n)A^{37}$. The threshold for this reaction is 1.60 Mev. Protons of energy up to 10 Mev are produced in condensed materials at the rate of about two per gram per day at sea level by the nucleonic component of cosmic radiation. These cosmic ray protons may be expected to produce an amount of A^{37} in the drums in the same order of magnitude as the sensitivity of these experiments.

[6] K. Way and E. Wigner, Phys. Rev. **73**, 1318 (1948).

In order to measure the cosmic ray background effect a 200 liter drum was exposed for a period of 62 days at the top of Mt. Evans, Colorado (14 150 feet elevation) through the courtesy of the Inter-University High Altitude Laboratory. It was shipped back to the Brookhaven laboratory and processed. The saturation A^{37} activity in this drum was 3.5 ± 0.5 disintegrations per minute and the activity was found to decay with the correct half-life. The corresponding cosmic-ray production rate for A^{37} in carbon tetrachloride was therefore $(1.1\pm0.2)\times10^{-5}$ disintegrations per minute per gram at atmospheric depth of 610 grams per cm^2. Assuming that the A^{37} observed was produced by the nucleonic component which has an exponential absorption mean free path of 140 grams per cm^2, the saturation activity expected in a 200 liter drum at sea level should be 0.17 disintegration per minute. The observed activity was less than this by a factor of six. Therefore the cosmic ray component producing A^{37} in the drum has an average absorption mean free path of less than 90 grams per cm^2.

An experiment was performed with a 3900 liter container located near the Brookhaven reactor as described. The saturation A^{37} activity observed was 1.23 ± 0.06 disintegrations per minute. The decay of this activity was followed and found to have the proper half life of 33 ± 3 days. This corresponds to a cross section of $(2.1\pm0.1)\times10^{-42}$ cm^2/atom for the antineutrino capture cross section (see Table I). However, it is probable that this activity was cosmic ray induced; the A^{37} production rate at sea level is $(2.0\pm0.1)\times10^{-7}$ disintegration per minute per gram of carbon tetrachloride uncorrected for shielding effects. The tank was partially shielded by the reactor since the tank stands one foot away from the shield of the reactor and the shield extends about twelve feet higher than the tank. The tank is eleven feet high and four feet in diameter, hence the lower portions of the tank were shielded by the upper portions of the tank. An approximate calculation of the shielding factor leads to a corrected A^{37} production rate of about 6×10^{-7} disintegration per minute per gram at sea level (atmospheric depth of 1035 grams per cm^2). The amount of A^{37} observed in the 3900-liter tank near the reactor, corrected for shielding, would correspond to a saturation activity of 0.20 disintegration per minute in a 200-liter container. This value is over six times greater than the upper limit of 0.03 disintegration per minute formed with an unshielded 200-liter container irradiated in the laboratory away from the reactor. Further experiments are to be performed to examine this point. At present it is concluded from our experiments that the cross section for the $Cl^{37}(\bar{\nu},e^-)A^{37}$ reaction is less than 2×10^{-42} cm²/atom for fission product antineutrinos.

An experiment was performed with the 3900-liter tank buried under 19 feet of earth (970 grams/cm²) to explore the possibility of producing A^{37} in the tank by processes other than those associated with the nucleonic component of cosmic radiation. This amount of earth will reduce the intensity of the nucleonic component by a factor of about one thousand. The amount of A^{37} activity in the buried tank was found to be less than 0.05 disintegration per minute at saturation, a limit set by the sensitivity of the counter used. The limit corresponds to detecting 70 neutrino captures per day in the 3900-liter tank. Since the 3900-liter tank with its associated counters was more sensitive for detecting neutrinos than any previously reported device, it is of interest to consider the possibility of detecting neutrinos from the sun. Two processes are now considered important for energy production in the sun, the proton-proton chain and the carbon-nitrogen cycle.[7] The neutrinos from the proton-proton chain have a maximum energy (0.41 Mev) below the threshold of 0.816 Mev for the $Cl^{37}(\nu,e^-)A^{37}$ reaction and therefore neutrinos from this chain could not be detected by this method. The neutrinos in the carbon-nitrogen cycle arise from the positron decays of N^{13} and O^{15} which have maximum neutrino energies of 1.24 and 1.68 Mev respectively. Based upon this cycle, the flux of neutrinos at the surface of the earth is approximately 6×10^{10} neutrinos/cm² sec.[8] The estimated average cross section for these neutrinos is about 3×10^{-46} cm² for the reaction. The experiment with the buried 3900-liter tank sets an upper limit on the neutrino flux from the sun of 1×10^{14} neutrinos/cm² sec if the energy production of the sun is entirely through the carbon-nitrogen cycle.

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[7] E. E. Salpeter, Ann. Rev. Nuc. Sci. **2**, 41 (1953).
[8] F. G. Houtermans and W. Thirring, Helv. Phys. Acta **27**, 81 (1954).

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Interactions of the Heavy Nuclei of the Cosmic Radiation*

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Fragmentation probabilities have been obtained for the interactions of heavy nuclei with target nuclei of photographic emulsion and a light element absorber (gelatin and cellulose acetate). The predictions of a simple geometrical model for the fragmentation probabilities in light elements derived from those observed in emulsion are in agreement with experimental observations. Measurements of the interaction mean free paths of heavy nuclei at different latitudes indicate that the interaction cross section is energy insensitive and is smaller than geometric. Curves are presented showing the ratio of the secondary light element (Li, Be, B) flux relative to the medium element ($6\leqslant Z\leqslant 10$) flux as a function of atmospheric depth.

INTRODUCTION

THE heavy primary component of the cosmic radiation consists of nuclei with charges ranging from $Z=3$ to $Z\sim26$.[1] The flux of these nuclei is small in comparison to the proton and α-particle flux (~1 percent of the proton flux) and increased statistics can be obtained by studying the behavior of various charge groups rather than that of individual charged nuclei. In the following discussion three different charge groups will be referred to: (1) L, light nuclei, $3\leqslant Z\leqslant5$, (2) M, medium nuclei, $6\leqslant Z\leqslant10$, and (3) H, heavy nuclei, $Z>10$.

In its passage through the atmosphere the heavy primary component changes in character since some particles are lost from the beam because of interaction and ionization loss while others are added to the beam as fragmentation products of interactions of nuclei of higher charge. Since the interaction mean free path of heavy nuclei in atmosphere is quite short (~20 to 40 g/cm²) direct measurements on the chemical composition must be made at extremely high altitudes. At the balloon altitudes attained on most experiments dealing with the heavy nuclear component of the cosmic radia-

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[1] For a review of the situation as of 1951 see the article by B. Peters in *Progress in Cosmic Ray Physics*, edited by J. G. Wilson (North Holland Publishing Company, Amsterdam, 1952), Vol. I. A survey covering results as of the fall of 1953 is given by E. P. Ney in the Report of the Duke Conference on Cosmic Radiation (unpublished).