Reports

Solar Neutrinos: Proposal for a New Test

Abstract. *The predicted flux on the earth of solar neutrinos has eluded detection, confounding current ideas of solar energy production by nuclear fusion. The dominant low-energy component of that flux can be detected by mass-spectrometric assay of the induced tiny concentration of 1.6 × 10^7 year lead-205 in old thallium minerals. Comments are solicited from those in all relevant disciplines.*

Detection and measurement of the flux on the earth of solar neutrinos (6 × 10^{10} cm^{-2} sec^{-1}) appears to be the only definitive test of current models of solar energy production, which postulate thermal fusion of light nuclei (hydrogen burning) in the solar core. A recent review by Bahcall and Davis (*1*) summarizes the essentially negative results of the experiment of Davis and co-workers: for the last 5 years the average yield of ^{37}Ar from neutrino capture in ^{37}Cl has been only about one-sixth of the rate predicted for this nucleus by almost all solar models, and within twice the experimental uncertainty the rate is not certainly above the estimated background. However, because of the rather high neutrino energy threshold for capture in ^{37}Cl, 0.82 Mev, the predicted capture rate excludes capture of about 95 percent of the solar neutrino spectrum. In fact, about 80 percent of the neutrinos expected to be captured in ^{37}Cl are those of very low relative intensity (~ 10^{-4} of the total solar neutrinos emitted) and very high energy ($E_{\max}$ = 14 Mev) that come from the rare decays of ^{8}B formed in a weak branch of the solar fusion chain. In perhaps the least radical (*2, 3*) of many proposed revisions (*1–6*) of the solar model or of the relevant nuclear or neutrino physics it is predicted that only this small flux component is overestimated in the solar model calculations, which lowers the predicted rate into near agreement with the observed value. Even if such revisions were tenable, the ^{37}Cl value is so low that it challenges the basic ideas (*1*) that solar neutrinos do reach the earth, or leave the sun, or are now being produced. Clearly, progress toward understanding the solar energy mechanism is now blocked pending definitive evidence as to the presence of the expected bulk of *low-energy* solar neutrinos.

This interest has stimulated renewed inquiry into alternate neutrino capture reactions with low energy thresholds, and Davis and co-workers have advanced the feasibility of experiments with ^{7}Li and ^{71}Ga targets in pilot tests (*1*). These experiments should yield of the order of one neutrino capture product atom per month per ton of target of a short-lived radioactive species, which can be extracted chemically and detected with good efficiency by following its decay, as for ^{37}Ar. Usable sensitivity requires massive targets—for example, 40 tons of Ga.

The availability of other methods of detecting modest numbers of neutrino capture product atoms, such as mass spectrometry or laser-excited resonance fluorescence, brings into consideration many capture reactions whose products are too long-lived for decay counting. With a longer half-life of the product atom, a larger concentration of product atoms can build up in the target mass. Thus a much higher overall sensitivity in terms of reduced target mass requirements can be realized, compared to the "decay counting" experiments, despite the lower detection efficiency of these other methods.

We have examined all possible candidate neutrino capture (inverse electron capture) reactions, imposing the requirements of (i) low energy threshold, ⩽ 0.4 Mev, to gain sensitivity to the intense neutrinos produced in the proton-proton ($p + p$) reaction (*1*); (ii) adequate and predictable cross section; (iii) very long product half-life, so that the product would have accumulated and have been retained in a suitable target mineral over geologic time, but less than 10^8 years so that it would not have survived from primordial generation; and (iv) tolerable yields of competing reactions giving the same product in situ. These requirements are satisfied only by the reaction

$$^{205}\text{Tl} + \nu\ (E_\nu \geq 46\ \text{kev}) \rightarrow\ ^{205\text{m}}\text{Pb}\ (E_{\text{exc}} = 2.3\ \text{kev}) + e^-$$

where ν is a neutrino, E_ν is the neutrino energy, E_{exc} is the energy of the excited $^{205\text{m}}$Pb nucleus, and e^- is an electron. The neutrino capture to the fortuitously low-lying excited state, $^{205\text{m}}$Pb, is followed by its fast isomeric decay to the ^{205}Pb ground state, whose 16-million-year half-life for $L + M + N$ . . . electron capture effectively traps the neutrino capture event. Direct neutrino capture to the ^{205}Pb ground state is relatively insignificant (~ 10^{-5} of that to the $^{205\text{m}}$Pb excited state).

The cross-section calculations (*7*) for neutrino capture are based on the log *ft* value (5.3 ± 0.1) for the inverse reaction, orbital electron capture decay of the $^{205\text{m}}$Pb state. (Log *ft*, where *f* is a function of decay energy and *t* is the half-life, is a measure of the probability of electron capture, and, hence, of neutrino capture.) This decay has an unmeasurably small branching ratio (10^{-13}); its log *ft* value is reliably estimated by analogy with all three neighboring transitions connecting the same single particle states with log *ft* values of 5.1 or 5.2. The predicted neutrino capture rate is 430 ± 100 solar neutrino units [SNU (*1*)] (~ 85 percent arising from the low-energy $p + p$ neutrinos) compared to 5.6 SNU predicted for ^{37}Cl.

In the most available of the very rare occurrences of known thallium crystalline minerals, lorandite (TlAsS$_2$), of estimated age ~ 10 million years, we predict a trapped ^{205}Pb concentration of 132 atoms per gram of lorandite, arising from solar neutrino capture. We are engaged in a detailed study of the feasibility of a solar neutrino flux measurement using a few kilograms of lorandite per sample. Our current considerations on procedure follow.

The methods considered for assay of the ^{205}Pb impose severe tolerance limits on the natural lead abundance in the sample, inasmuch as the tiny amount of ^{205}Pb must be distinguishable from the macroscopic amounts of the stable lead isotopes, masses 204, 206, 207, and 208, that will surely be present. The best anticipated ability of a mass spectrometer to detect mass 205 in the presence of the scattering tail from ~ 10^{10} times as much mass 206 translates into a maximum allowable lead impurity in the mineral of a few parts per million (ppm); even this limit requires inclusion in the Pb-Tl separation procedure of a stage of isotopic enrichment of mass 205 by 10^4, at a cost of 90 percent loss of the ^{205}Pb.

Thallium minerals are very rare; only lorandite appears to be available in amounts exceeding a few grams, and that only at the site of its original discovery (*8*) as a minor constituent in an arsenic sulfide ore deposit in southern Macedonia, Yugoslavia. There it is believed to exist in adequate abundance at a depth of about 120 m (*9*). We have measured the Pb contamination of a museum specimen (*10*) from this source as ~ 3 ppm, usable for the neutrino experiment. The estimated age (*9*) of the ore is 10 million to 15 million years.

A serious question exists as to whether the depth of the ore is adequate to provide sufficient shielding against the high energy cosmic-ray muons. These may produce protons in deep rock which can participate in the ^{205}Tl(p,n)^{205}Pb reaction in the lorandite, giving the major contribution of background ^{205}Pb, just as the muon-generated ^{37}Cl(p,n)^{37}Ar reaction gives the principal background in Davis's experiment even at his target depth of 1480 m. We have crudely estimated the depth of burial of lorandite at which the muon-generated background would be tolerable as 300 m. (We are now measuring these cross sections with Davis in the muon beam at Fermilab.) The estimated 300-m depth requirement is much greater than the 120-m depth of the mine shaft in which the Macedonian deposit was found, and it is not yet known if that deposit extends much deeper. However, we have been advised by consultant geologists (*11*) that the rate of erosion of rock overburden at the site of the deposit is estimated to have been at least 200 to 300 m within at most the last million years, so that lorandite now 120 m deep was 300 to 400 m deep a million years ago, and perhaps even deeper earlier. Thus the known deposit was probably fairly well shielded over most of the last mean life of ^{205}Pb, and so is useful for the experiment. It should be possible to refine the depth requirements with the Fermilab experimental results and studies of past erosion rates at the Macedonian site. Lorandite has also been identified in milligram amounts at mine sites in western states (*12*). We are, of course, interested to learn of other sources of crystalline thallium minerals.

The separation and enrichment procedure for lorandite would proceed roughly as follows. Each 3-kg sample is subjected to a sequence of stages of initial chemical separation in which Tl is removed to a residue of ~ 1 mg; the original Pb content, ~ 10 mg, remains. Electromagnetic separation is then used to enrich mass 205, and finally a series of ultrapure liquid-liquid chromatographic separation stages should provide ~ 60-ng samples each containing $\sim 5 \times 10^4$ atoms of ^{205}Pb, $\sim 2 \times 10^{14}$ atoms of ^{206}Pb, and a ratio ^{205}Tl/^{205}Pb $\approx$ 10 corresponding to a Tl reduction factor of 10^{17}. If the ^{205}Pb assay can employ laser-excited resonance fluorescence source ionization and/or ion-beam detection in the mass spectrometry, the requirements for Tl stripping are much less severe, perhaps by 10^8. There is a backlog of experience at Argonne in such techniques in transplutonic element chemistry at the level of a few atoms from massive targets (*13*).

The samples are then assayed for absolute ^{205}Pb content to 10 to 20 percent accuracy by mass spectrometry using the isotope dilution method. The Argonne 100-inch double focusing mass spectrometer (*14*) attains more than 90 percent transmission efficiency at a resolution of 5000, sufficient to discriminate against the organic mass-205 peak. With an added second magnetic stage and ion retarding lens the scattered background tail from ^{206}Pb at mass 205 will not exceed 10^{-10} of the peak intensity of mass 206. We need 1 to 10 percent ionization efficiency, which we have attained with a surface ionization filament or gas magnetron source. With the former we can tolerate a ^{205}Tl/^{205}Pb ratio of > 10 because of differential volatility. If we can produce Pb ions at 1 percent efficiency using two-stage laser excited resonance fluorescence, the tolerance level for Tl in the mass spectrometer sample will be raised by many orders of magnitude, with a corresponding easing of the burden on the chemical purification.

Corrections are then made for various sources of "background"—that is, for the estimated ^{205}Pb content of lorandite arising from nuclear reactions of Tl and of Pb, Hg, and Bi impurities induced by alpha particle, proton, and neutron irradiations from natural radioactivities in the mineral's environment and from cosmic-ray muons. In view of the low yield from neutrino capture, each such other reaction is clearly a serious hazard, and each must separately be proved negligible. Preliminary estimates show that each is tolerable.

The age of lorandite mineralization is determined by dating techniques: on the lorandite directly by counting spontaneous-fission tracks, if possible, or by the K-Ar method; on associated minerals in the deposit by these methods and/or by U-Pb measurements; and by standard geological stratification analysis.

From the corrected ^{205}Pb concentration observed, and the measured age of the Tl mineral, one obtains the neutrino flux averaged over that age, or, for older minerals, over a time span of the order of 23 million years, the mean life of ^{205}Pb. A result in reasonable agreement with current predictions for the major low-energy part of the solar neutrino spectrum would be especially interesting to compare with the data from the proposed ^{71}Ga experiment (*1*), which is also sensitive to the low-energy neutrinos, but which gives an essentially current view of the flux because of the 11-day life of the ^{71}Ge product of neutrino capture. This comparison could speak to the validity of recent solar model studies (*4*, *5*) that suggest possible gross fluctuations or cyclic behavior in the thermal fusion (and hence neutrino emission) rate, with periods of the order of 1 million to 2 million years. Such oscillations would be severely damped out in respect to light radiation from the photosphere, so evidence of their occurrence would be suppressed. If the long-time average solar energy production is indeed due to hydrogen burning, and if the emitted neutrinos reach the earth, the ^{205}Tl detector should show an average neutrino flux value equal to that of the steady-state models, for the several cycles of possible fluctuation over which it integrates, whereas the ^{71}Ga and ^{37}Cl detectors may now be looking at a trough in the neutrino rate.

Estimation of the uncertainties accruing to this measurement—those of the neutrino absorption cross section, the ^{205}Pb background, and the analysis and age determination—as they appear at present yields an overall uncertainty of perhaps 30 to 40 percent for the neutrino flux evaluation. While this can hardly be regarded as precisely testing the current solar model, what is now sorely needed is a resolution of the dilemma as to whether solar neutrinos are indeed reaching the earth, and with an intensity consistent with the model predictions. Should the answer given by this measurement be affirmative, further refinements can yield significantly improved accuracy.

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Aluminum-26 in Deep-Sea Sediment

Abstract. *The activity of ^{26}Al in a North Pacific core was found to be 0.081 ± 0.046 disintegration per minute per kilogram of dry sediment, which corresponds to a ^{26}Al/^{10}Be ratio of 0.018 ± 0.011 (or 0.019 ± 0.012 when this ratio is corrected for decay). This ratio is in good agreement with that measured in Greenland ice, 0.017 ± 0.008. These ratios are also in agreement with the calculated values for the production of these isotopes by cosmic rays in the atmosphere: 0.013 ± 0.006. The contribution of cosmic dust bearing ^{26}Al seems small in comparison with the production of this nuclide in the atmosphere.*

Bombardment of atmospheric constituents by galactic cosmic rays produces two radionuclides that have million-year half-lives ($T_{1/2}$), ^{10}Be ($T_{1/2} = 1.5 \times 10^6$ years) and ^{26}Al ($T_{1/2} = 0.716 \times 10^6$ years) (*1*). The influx of cosmic dust can also bring to the earth some cosmogenic radionuclides such as ^{26}Al, ^{53}Mn ($T_{1/2} = 3.7 \times 10^6$ years), and ^{59}Ni ($T_{1/2} = 0.080 \times 10^6$ years), which are produced principally by the interaction of solar protons and α particles with cosmic dust (*2*). Pelagic sediments and polar ices are considered to be the best materials in which to detect these radionuclides (*1, 2*).

The measurement of such nuclides in marine sediments can furnish useful information about oceanic sedimentation rates and cosmic dust accretion rates in the last millions of years. It has been proposed that simultaneous measurements of ^{26}Al and ^{10}Be would be a good way to acquire such information (*3*).

Amin *et al.* (*4*) found the mean specific activities of ^{26}Al and ^{10}Be in two cores from the Pacific basin to be 0.5 and 4 disintegrations per minute (dpm) per kilogram of dry sediment, respectively, which correspond to an average ^{26}Al/^{10}Be ratio of 0.12 and imply that the ^{26}Al content is higher by an order of magnitude than that expected from the spallation of atmospheric argon. Indeed, Lal and Peters (*5*) estimated the global production rates of ^{26}Al and ^{10}Be by spallation of atmospheric nuclei to be 8×10^{-9} and 8×10^{-7} dpm cm^{-2} year^{-1}, respectively, which corresponds to a ^{26}Al/^{10}Be ratio of 0.01. The high ^{26}Al content found by Amin *et al.* in the Pacific sediments was interpreted as a consequence of a considerable influx with cosmic dust (*6*). Wasson *et al.* (*7*) reported a ^{26}Al content of 0.8 dpm per kilogram of dry sediment. This high ^{26}Al content was also interpreted as a consequence of ^{26}Al influx with cosmic dust, although one cannot completely rule out the possibility that argon spallation is responsible for all the observed activity. McCorkell *et al.* (*8*), however, found that the ^{26}Al/^{10}Be ratio in Greenland ice was 0.017, which is an order of magnitude lower than the ratio obtained by Amin *et al.* (*4*). This low ratio can be explained as resulting entirely from spallation of atmospheric nuclei without the need to postulate a ^{26}Al influx with cosmic dust. Tanaka *et al.* (*9*) measured the mean specific activities of ^{26}Al and ^{10}Be in a core from the Pacific basin to be 0.02 ± 0.26 (*10*) and 4.4 ± 0.9 dpm per kilogram of dry sediment, respectively, which results in an average ^{26}Al/^{10}Be ratio of less than 0.06, the upper limit of the ^{26}Al activity being 0.28 dpm per kilogram.

In the past ^{26}Al in sediments was detected by measurement of coincidence events of two annihilation γ-rays (0.511 Mev), because the small NaI(Tl) crystals available then (about 7.5 by 7.5 cm) were extremely inefficient detectors of 1.8-Mev photons. This peak at 0.511-0.511 Mev is common for all positron (β^+) emitters such as ^{22}Na and ^{44}Ti and not necessarily specific for ^{26}Al. Perhaps remeasurements of the ^{26}Al contents of earlier samples in a more specific way with more sensitive instruments will contribute to the settlement of the question.

It was for this reason that we undertook remeasurements of the samples measured by Tanaka *et al.* (*9*). The samples are Al_2O_3 extracted from half (in vertical cutting) of a red clay core, 7.8 cm in diameter and 1 m long, which came from a depth of 5439 m at 23°07′N, 135°45′E. The Al_2O_3 content was vertically uniform throughout the core: 16.38 ± 0.32 percent (dry weight). The method of extraction and purification of aluminum has been described in (*9*). The chemical yield of aluminum was 72.2 ± 2.8 percent. The core was cut in four sections, each about 25 cm long. For the present mea-

Table 1. Measurements of ^{26}Al in an Al_2O_3 sample from Pacific sediment.

Energy (Mev)	Nuclide and efficiency	Count rate (10^{-4} count min^{-1})				Activity* (dpm per kilogram of dry sediment)
		Gross	Contribution		Net	
			Background	Thorium†		
			At Gif-sur-Yvette			
0.511-2.34	^{26}Al, 6%	423 ± 22	424 ± 18	0 ± 2	−1 ± 28	0.00 ± 0.14
			At Issy-les-Moulineaux			
0.511-2.34	^{26}Al, 5.6%	168.1 ± 6.8	152.6 ± 5.2	0 ± 1.6	15.5 ± 8.7	0.081 ± 0.046
0.511-1.81	^{26}Al, 1.9%	265.3 ± 8.6	257.6 ± 8.8	0 ± 0.7	7.7 ± 12.3	0.12 ± 0.19
0.511-0.511	^{26}Al (β^+), 6%	730 ± 14	729 ± 15	0 ± 2	1 ± 21	0.00 ± 0.10
0.583-2.61	^{232}Th(^{208}Tl), 1.5%	769 ± 15	772 ± 12		−3 ± 19	

*The errors include all known sources of errors: counting statistics (1 standard deviation), counting efficiency (± 5 percent), and chemical yield (± 4 percent). †Contributions of thorium to the peaks at 0.51-2.34 Mev, 0.511-1.81 Mev, and 0.511-0.511 Mev are 8.5, 4.0, and 8.4 percent, respectively, of the count rate in the peak at 0.583-2.61 Mev.