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Radiochemical solar neutrino experiments, "successful and otherwise"

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Abstract. Over the years, several different radiochemical systems have been proposed as solar neutrino detectors. Of these, two achieved operating status and obtained important results that helped to define the current field of neutrino physics: the first solar-neutrino experiment, the Chlorine Detector (^{37}Cl) that was developed by chemist Raymond Davis and colleagues at the Homestake Mine, and the subsequent Gallium (^{71}Ga) Detectors that were operated by (a) the SAGE collaboration at the Baksan Laboratory and (b) the GALLEX/GNO collaborations at the Gran Sasso National Laboratory. These experiments have been extensively discussed in the literature and in many previous International Neutrino Conferences. In this paper, I present important updates to the results from SAGE and GALLEX/GNO. I also review the principles of the radiochemical detectors and briefly describe several different detectors that have been proposed. In light of the well-known successes that have been subsequently obtained by real-time neutrino detectors such as Kamiokande, Super-Kamiokande, SNO, and KamLAND, I do not anticipate that any new radiochemical neutrino detectors will be built. At present, only SAGE is still operating; the Chlorine and GNO radiochemical detectors have been decommissioned and dismantled.

1. Introduction

The Standard Solar Model (SSM) that was put forth by John Bahcall and colleagues [1] is based on the concept that solar energy is the product of nuclear reactions that convert hydrogen in the Sun into helium, releasing 26 MeV of energy and producing isotopes of the chemical elements He, Li, Be, and B, and to a lesser extent, C, N, and O. In this process, electron-flavor neutrinos, ν_e, are emitted in beta-decay processes. These reactions occur in a step-wise manner because the temperature in the solar core is low, ~15 million degrees Kelvin or an energy of ~ 1 keV. Figure 1 shows the calculated energy spectra and fluxes of the solar ν_e produced by these reactions, from the SSM of Bahcall and Pinsonneault [2].
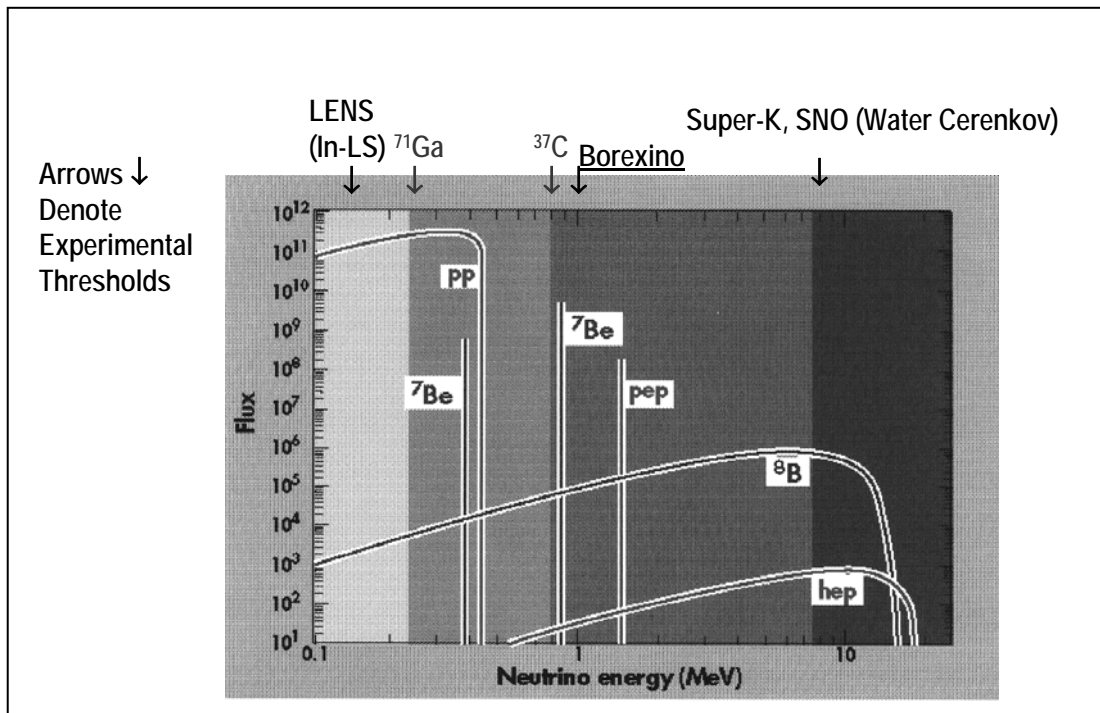
Figure 1. Predicted Energy Spectra of Solar Neutrinos from the Standard Solar Model [2}. Energy thresholds of several neutrino detectors are denoted by ↓.

The early stages in the development of solar neutrino detection were primarily dominated by radiochemical methods. Out of the four neutrino detectors that were operating by the early 1990's, three were radiochemical. The first experiment was the radiochemical Chlorine (^{37}Cl) detector that was developed and successfully operated by Ray Davis and colleagues in the Homestake Mine in the USA [3]. Its goal was to detect the solar neutrinos that were predicted by the SSM. Davis impressively succeeded in achieving this goal, thereby winning the Nobel Prize in Physics in 2002. Perhaps more important for the field of neutrino physics was the fact that he also uncovered an *unexpected result*, the famous Solar Neutrino Problem (SNP): the flux of detected solar neutrinos was much less than that predicted by the SSM. Although there were doubters who initially thought that the Davis experimental result was wrong, many others worked to develop follow-on experiments to solve the SNP and eventually did so.

The second solar neutrino experiment to be operated was the real-time Kamiokande detector in Japan [4] that was originally built to search for proton decay but was converted to solar-neutrino astronomy after it detected elastic scattering of neutrinos emitted by Super-Nova 1987A. The third and fourth solar neutrino experiments were the two variations of the Gallium (^{71}Ga) radiochemical detectors, SAGE [5] and GALLEX [6], which subsequently was modified and run as GNO. As is very well known, all of these experiments succeeded in verifying the existence of the solar neutrino deficit.

2. Radiochemical detection of solar neutrinos

2.1. Principles of radiochemical detection

The radiochemical detection method relies on neutrino capture via the Weak Interaction (also called Inverse Beta Decay, IBD) in a target chemical element with (A, Z),

$$\nu_e + (A, Z) \rightarrow e^- + (A, Z+1)^*.$$
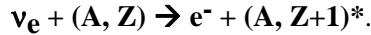
The target element is transmuted into another chemical element, (A, Z+1)*, where the * denotes that the product nuclide is radioactive with half-life $T_{1/2}$.

The detection method requires clever sensitive radiochemical methods to separate and remove the few atoms of product element (Z+1) from the target Z. Because the cross section for neutrino capture is very small, $<10^{-40}$ cm^2, the targets must be huge, containing many tons of material. The experiments are done deep underground to minimize cosmic-ray interactions in the target that can produce protons, because (p,n) reactions mimic ν_e capture.

Runs are done in a batch mode, with solar exposure times on the order of $2T_{1/2}$. Typically one isolates and collects ~10 product atoms from ~10^{30} target atoms with efficiency ≥90%. The product is purified from contaminants, especially radioactive ones such as U, Th, and Rn. In the Chlorine and Gallium experiments, the product atoms were converted to suitable gaseous chemical forms, ^{37}Ar and ^{71}GeH$_4$, respectively, that could be placed within high-efficiency, low-background gas-filled proportional counters. These radioactive product nuclides were identified unequivocally by their energy spectra and their half-lives. Note that for a radioactive product nuclide, the measured log-ft value for β decay from the ground state of (A, Z+1)* to (A, Z) leads via the Principle of Microscopic Reversibility to the value of the cross section for ν_e capture on (A, Z), for the ground-state-to-ground-state transition.

2.2. Radiochemical neutrino detectors that have been proposed

In chapter 13 of his monograph, Bahcall [1] described seven different radiochemical detectors that have been proposed and researched over the years. Their individual histories are fascinating. Table 1 summarizes important features of each of these detectors.

Table 1. Radiochemical solar neutrino detectors

Neutrino capture on target	$T_{1//2}$	Energy threshold	Comments
Chlorine, ^{37}Cl → ^{37}Ar	35.0 days	0.814 MeV	"successful"
Gallium, ^{71}Ga → ^{71}Ge	11.4 days	0.233 MeV	"successful"
Iodine, ^{127}I → ^{127}Xe	36 days	0.789 MeV	prototype only
Molybdenum, ^{98}Mo → ^{98}Tc	4×10^6 years	>1.74 MeV	"unsuccessful"
Lithium, ^{7}Li → ^{7}Be	53 days	0.862 MeV	R&D only
Bromine, ^{81}Br → ^{81}Kr	2×10^5 years	0.470 MeV	R&D only
Tantalum, ^{205}Tl → ^{205}Pb	14×10^6 years	0.054 MeV	R&D only

The Chlorine detector stopped operations several years and its final results have been reported extensively. The two Gallium detectors continued to run into this decade. Recent results from SAGE and GALLEX- GNO will be presented updated in Section 3 below.

Here I mention some details about two of the other radiochemical detectors listed in Table 1.

The ^{127}I detector was developed by K. Lande et al. [7]. Its threshold was very similar to that of the ^{37}Cl detector, and its purpose was to provide an independent check of the SNP neutrino deficit from ^{37}Cl. The radiochemistry used was analogous to the Chlorine experiment; neutrino capture on the halogen target produced a noble gas that could be easily removed from the liquid target. The researchers built a prototype and developed novel automated chemical steps to segregate the product ^{127}Xe atoms into day and night fractions. Prototype testing was ended a few years ago when a business decision was made to close down the Homestake Mine.

Another radiochemical detector based on ^{98}Mo was developed by K. Wolfsberg et al. [8] to check the Chlorine detector's neutrino deficit over geological times. A very deep molybdenum-sulfide ore body was selected that was adequately shielded from cosmic rays. It was chemically processed with a novel chemical system to separate Tc from Mo at the industrial chemical smelter where the molybdenum sulfide was roasted to molybdenum oxide. They did an actual run with the deep ore and did detect ^{98}Tc. However, the smelter equipment had been contaminated with cosmogenic Tc from previously processed Mo from shallow ore deposits. Their measured neutrino production rate was several times larger than the SSM value, a clear sign of ^{98}Tc contamination. Adequate funding was never obtained to redo their experiment with properly cleaned equipment at the smelter, so the unsuccessful project was aborted.

3. Results from the Gallium radiochemical neutrino-detectors

3.1. Recent solar results from SAGE

The range of rates predicted for ^{71}Ga by the SSM are: (a) 122-131 SNU without neutrino oscillations, and (b) 68-72 SNU (from a global fit to neutrino data) with neutrino oscillations.
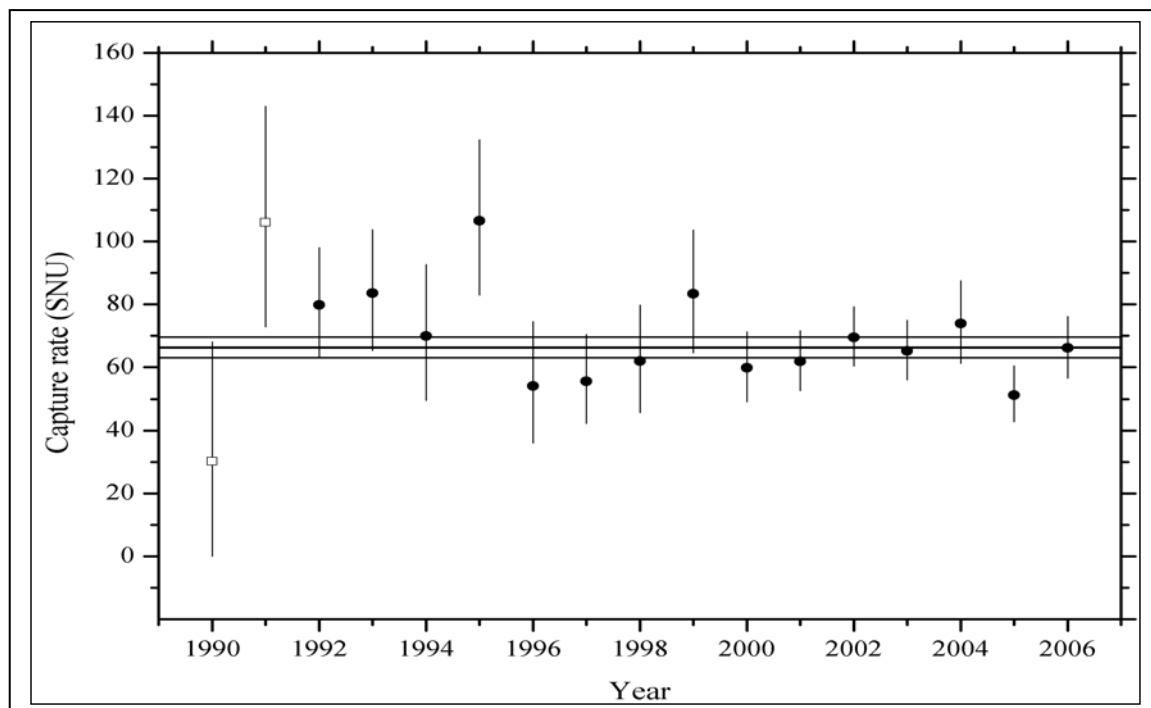

Figure 2. SAGE results. 1 SNU = 1 neutrino capture per sec per 10^{36} target atoms.

Results from SAGE [9], taken from 157 runs for the seventeen-year running period from1990-2006, are shown in Figure 2. These data are averaged in one-year bins. The mean value extracted from these **SAGE** data is 66.2 + stat. (+3.3 –3.2) + syst. (+3.5 – 3.2) SNU, or **66.2 $^{+4.8}_{-4.5}$ SNU**.

Gavrin has pointed out that at present, SAGE is the only running experiment that has a sufficiently low threshold to be sensitive to *pp* neutrinos as well as to ^{7}Be and higher-energy solar neutrinos. SAGE plans to continue running for >3 more years while Borexino is measuring solar ^{7}Be neutrinos, to make a direct comparison of these two low-energy neutrino experiments.

3.2. Recent solar results from GALLEX-GNO

After GNO stopped running, scientists from GALLEX and GNO have recently done a complete reanalysis of the GALLEX data set [10, 11]. They began by recalibrating each gas-proportional counter with ~10^5 injected ^{71}Ge decays, which they could not have done before completing the low-rate solar runs for fear of contaminating the counters. They then did a Pulse Shape Analysis (PSA) of the individual counts, instead of the previously used less precise Rise Time Analysis (RTA). They also improved the efficiency of the Rn cut and of the background determinations.

Figure 3 shows the revised GALLEX values as black dots (for each of the four run periods, the point on the left is the old RTA value, the point to the right is the new PSA value) and the three GNO values as gray dots. The new PSA method improves the accord between the GALLEX period-3 value and the other periods. The revised values are: GALLEX combined for periods 1-4, 73.4 + stat. (+6.1 – 6.0) + syst. (+3.7 – 4.1) SNU; GNO combined, 62.9 + stat. (+5.5 – 5.3) ± syst.2.5 SNU; and the **combined new GALLEX + GNO result, 67.6 ± 4.0 stat. ± 3.2 syst. SNU.**
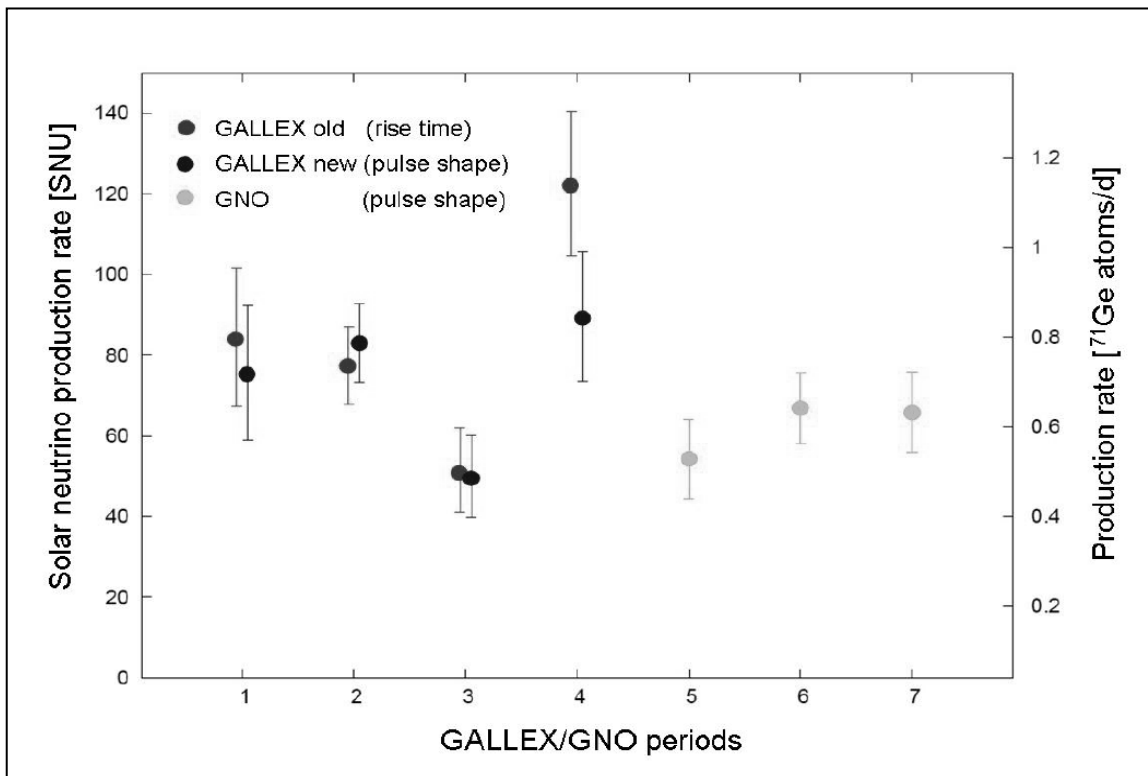

Figure 3. Revised results from GALLEX and GNO.

Note that GALLEX-GNO ended operations in March 2006, after 15 years of operation. The experiment has been decommissioned and the gallium removed from the Gran Sasso Laboratory.

3.3. Comparison of SAGE and GALLEX-GNO results

There is impressive agreement between the results of these two experiments, 66.2 vs. 67.6 SNU for SAGE and GALLEX-GNO, respectively, with overall errors ~5 SNU for each experiment. Both experiments have reached the point where their statistical errors roughly equal their systematic errors. These results provide clear evidence for solar neutrino oscillations at low energies, <1 MeV.

4. Exposure of radiochemical detectors to neutrinos from intense radioactive sources

GALLEX [12] and SAGE [13] are the only two neutrino detectors to have been "exposed to" (some people have said "calibrated by") known fluxes of neutrinos emitted by intense "man-made" radioactive sources. The two radionuclides used in this application are ^{51}Cr by GALLEX and by SAGE, and ^{37}Ar by SAGE. The measured disintegration rates of these isotopes, both of which decay by electron capture with known decay schemes, directly gives the sources' experimentally determined neutrino emission rates. The source strength has to be large enough, ~MCi, so that the solar production rate of ^{71}Ge in the gallium target becomes a small background in the source exposure. I emphasize that each source exposure was a major experiment in its own right, comparable in complexity and difficulty to the radiochemical solar neutrino experiment.

There were two different GALLEX Cr-source experiments. Their results, expressed as the ratio, R, of the measured ^{71}Ge rate to the ^{71}Ge rate expected from the known ^{51}Cr-source decay rate, have recently been reanalyzed by the PSA method, which was discussed in Section 3.2. The mean R-value from the two source experiments had previously been reported as 93 ± 8% (1σ error). With the PSA, the reevaluated GALLEX mean = 88.2 ± 7.8% (1σ), with the values from the individual exposures being R1 = 95.3 ± 11% and R2 = 81.2 ± 11%.

The corresponding SAGE results were R = 95 ± 12 % for the ^{51}Cr source, and 79 +(+9 –10)% for the ^{37}Ar source. All four of these values overlap within their errors, in a band that is centered at R = 88%. These values have physical significance in that they are all <1σ from the value of 95 ± 1% that is expected from the neutrino-capture transition only to the ground state of ^{71}Ge. There have been extensive discussions in the literature about the contribution of transitions to ^{71}Ge excited states, with a value of 5 ± 3% having been estimated by Bahcall. The four source results cited here imply that the excited-state contributions in ^{71}Ge are close to 0%.

5. Conclusions

The radiochemical Cl and Ga neutrino detectors formed the first generation of solar neutrino experiments. In fact, in the early 1990's, they and Kamiokande were the only operating neutrino experiments. They have been important contributors to the advances in our understanding of ν properties, and in identifying and solving the Solar Neutrino Problem.

However, the radiochemical experiments operate in batch mode, yielding only one physical quantity, the production (or SNU) rate, which is proportional to the solar ν flux, and which must be interpreted in terms of the SSM. On the other hand, the real-time neutrino detectors see the neutrino interactions event by event, and yield several neutrino parameters, such as the ν spatial distribution in the detector, the energy spectra, directionality, and even the oscillation pattern. Note that SNO, by detecting both the CC and NC interactions simultaneously in real time, provided definitive proof of neutrino flavor oscillations independent of the SSM.

In addition, the real-time ν detectors have made consistent progress in lowering their detection thresholds, from ~ 9 MeV for Kamiokande to 4-5 MeV for Super-Kamiokande and SNO, to ~1 MeV for KamLAND and ~0.3 MeV for Borexino. Other planned real-time detectors, such as LENS, CLEAN, and e-Bubble aim to lower their thresholds further to detect solar *pp* neutrinos.

In my career in nuclear chemistry and physics, I have witnessed a *natural progression in which nuclear chemical methods*, where one observes the radioactive products of nuclear interactions after the interactions have occurred, *have been supplanted by real-time detection* of diverse nuclear phenomena, not only of neutrino reactions but also for example of nuclear fission and of complex heavy-ion high-energy nuclear reactions, such as those at RHIC.

For these many reasons, **I do not see that much incentive exists for further development of radiochemical ν detectors; certainly none appear imminent.**

However, I do think that (nuclear) chemists will continue to play a significant role in ν research, e.g., a) in developing new detector systems, such as metal-loaded liquid scintillators and cryogenic detectors; b) in detecting and reducing the levels of radioactive contaminants, such as U, Th, Ra, Rn, K; c) in developing new radioactive neutrino calibration sources; d) in studying the long-term chemical interactions and compatibility of new detector substances with detector construction materials, such as the detector containment vessels; and e) in being concerned about chemical safety issues in large-scale neutrino experiments that are done underground.

I close by noting that the history of the Neutrinos/Nuclear-Chemistry Group at BNL has closely followed the above trends. The group, under the leadership of Ray Davis, pioneered radiochemical neutrino detectors. I became group leader when we joined the radiochemical GALLEX experiment. After twelve years at Gran Sasso, we changed directions and participated in real-time neutrino detection in SNO. At present, we are developing other new real-time neutrino detectors, at Daya Bay, SNO+, and Mini-LENS/LENS.

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