Radiochemical solar neutrino experiments

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Abstract.
Radiochemical experiments have been crucial to solar neutrino research. Even today, they provide
the only direct measurement of the rate of the proton-proton fusion reaction, \(p + p \rightarrow d + e^+ + \nu_e\),
which generates most of the Sun’s energy. We first give a little history of radiochemical solar neutrino
experiments with emphasis on the gallium experiment SAGE – the only currently operating detector of
this type. The combined result of all data from the Ga experiments is a capture rate of 67.6 \(\pm\) 3.7 SNU.
For comparison to theory, we use the calculated flux at the Sun from a standard solar model, take into
account neutrino propagation from the Sun to the Earth and the results of neutrino source experiments
with Ga, and obtain 67.3\(^{+3.0}_{-3.5}\) SNU. Using the data from all solar neutrino experiments we calculate
an electron neutrino \(pp\) flux of \(\phi_{pp} = (3.41^{+0.17}_{-0.77}) \times 10^{10}/(\text{cm}^2\cdot\text{s})\), which agrees well with the prediction from a
detailed solar model of \(\phi_{pp} = (3.30^{+0.13}_{-0.14}) \times 10^{10}/(\text{cm}^2\cdot\text{s})\). Four tests of the Ga experiments have been
carried out with very intense reactor-produced neutrino sources and the ratio of observed to calculated
rates is 0.88 \(\pm\) 0.05. One explanation for this unexpectedly low result is that the cross section for
neutrino capture by the two lowest-lying excited states in \(^{71}\)Ge has been overestimated. We end with
consideration of possible time variation in the Ga experiments and an enumeration of other possible
radiochemical experiments that might have been.

1. Introduction and a little history
Our knowledge of neutrinos from the Sun is based on seven experiments: Homestake, Kamiokande,
SAGE, Gallex, GNO, Super-Kamiokande and SNO. More than half of these are radiochemical
experiments.

The detection of neutrinos by use of the inverse \(\beta\) decay reaction was proposed 60 years ago by
Bruno Pontecorvo [1]. This method of detection, which is the basis for radiochemical experiments, has
played a fundamental role in solar neutrino investigation. The idea to use neutrino capture in \(^{37}\)Cl to
observe the “undetectable” new particle proposed by Wolfgang Pauli was brilliantly realized to observe
solar neutrinos by R. Davis and collaborators in the world-famous experiment at the Homestake Gold
Mine [2, 3, 4]. The \(^{37}\)Cl experiment was built 4200 m.w.e. (meters of water equivalent) underground
and began to collect data in 1967. Between 1970–1994, 108 extractions of Ar were made from a tank
that contained 615 tons of \(^{37}\)Cl. The number of \(^{37}\)Ar atoms collected in each run was measured in
a miniature proportional counter. The result for the first measured capture rate of solar neutrinos at
the Earth was 2.56 \(\pm\) 0.23 SNU. The SNU unit (defined as 1 neutrino capture per day in a target that
contains 10\(^{36}\) atoms of the neutrino-absorbing isotope) was specially introduced by John Bahcall, who
had a fundamental role in the funding of the Cl experiment and the interpretation of its results, and whose
contributions cannot be overestimated. Bahcall was the first to fully develop a solar model that included

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all the physical parameters needed to calculate the solar neutrino flux at the Earth. He worked tirelessly to refine his calculations and it was the robustness of his solar model that eventually led all people to understand the significance of the discrepancy between the result of the Cl experiment and standard solar model (SSM) predictions.

The discrepancy identified in the Cl experiment attracted the attention of a significant number of scientists and it soon became known as “the solar neutrino problem”. This problem continued to bother the mind of scientists for more than 30 years. Especially important was the confirmation of the discrepancy by the Kamiokande experiment [5], a real-time detector of solar neutrinos that used a completely different method of detection – electron scattering, and which began to collect data in 1987. As a result there were no doubts that the flux of neutrinos in the high-energy part of the solar neutrino spectrum was significantly less than the calculations of the SSM. Kamiokande, with an analysis threshold of 7 MeV, was sensitive only to the high-energy $^8\text{B}$ neutrinos and the Cl experiment, whose major response was from the superallowed analog state at an excitation energy of 5.0 MeV in $^{37}\text{Ar}$, was also mostly sensitive to the $^8\text{B}$ neutrinos. Another significant development during this time was the confirmation of the results of the Bahcall SSM by a solar model independently developed by Sylvaine Turck-Chièze and collaborators [6].

Despite many attempts, the combination of these two experiments could not be explained on the basis of solar physics; rather, many scientists began to believe that it was necessary to reject some of our old ideas about neutrino properties and to develop new ones. Conclusive evidence for this suggestion could be obtained by measuring the low-energy part of the solar neutrino spectrum, which is produced in reactions that provide the vast majority of the Sun’s energy, and whose flux can be well predicted from the measured solar luminosity combined with a simple solar model. The need for experiments sensitive to low-energy solar neutrinos was recognized shortly after the first results from the Cl experiment were announced and many people began to consider radiochemical experiments with low-energy sensitivity, such as those shown in Table 1.

**Table 1.** Radiochemical solar neutrino detectors considered in 1972 [7] The relative response is given to the various sources of solar neutrinos. The mass of target element is the number of tons required to yield 1 neutrino capture/day from the sum of the $pp$ and $pep$ reactions. The relative response and mass were calculated from the 1972 values of solar flux and used cross sections that neglected excited states.

| Target, Product | Relative response (%) | Mass (tons) |
|----------------|-----------------------|-------------|
| $^{87}\text{Rb}$, $^{87m}\text{Sr}$ | $pp$ $pep$ $^7\text{Be}$ $^8\text{B}$ CNO | 74 2 21 1 3 32 |
| $^{55}\text{Mn}$, $^{55}\text{Fe}$ | 67 3 25 1 3 420 |
| $^{71}\text{Ga}$, $^{71}\text{Ge}$ | 69 2 26 0 3 19 |
| $^7\text{Li}$, $^7\text{Be}$ | 0 18 15 51 16 17 |

From these possibilities attention focused on $^7\text{Li}$, proposed in 1969 by John Bahcall [8], and on $^{71}\text{Ga}$, proposed in 1965 by Vadim Kuzmin [9]. Because of its high capture rate, low energy threshold of 233 keV and favorable half-life of 11.4 days, a Ga experiment appeared to be a most attractive possibility. The main problems with the Ga experiment were the acquisition of several tens of tons of the expensive element gallium and the development of a nearly lossless procedure for the extraction and purification of $^{71}\text{Ge}$.

**2. The Ga experiment**

Laboratory research to develop a gallium experiment began approximately in 1975. In the United States this work took place at Brookhaven National Laboratory under the direction of Ray Davis with
participation of B. Cleveland, J. Evans, G. Friedlander, K. Rowley, R. Stoener from Brookhaven, and W. Frati and K. Lande from the University of Pennsylvania [10]. Methods were developed to extract germanium from liquid gallium metal and from a GaCl$_3$ solution. After a few years, this group achieved success in development of these methods and chose the method based on GaCl$_3$ solution. To carry out the experiment a collaboration was initiated with a group from the Max Planck Institute at Heidelberg. Despite repeated requests and favorable reviews, the Ga experiment was, however, not funded in the United States. Rather, a special subcommittee of the Nuclear Science Advisory Committee recommended that interested scientists associate themselves with groups in Western Europe and/or the Soviet Union. The western European group, called Gallex, had been formed by the Heidelberg group under the direction of Till Kirsten when it became apparent that the experiment would not be funded in the US.

In the Soviet Union, at the Institute for Nuclear Research, laboratory investigations to develop a gallium experiment began about the same time in 1975. It was initially based on a GaCl$_3$ solution, but when it was learned that Soviet industry could not provide the necessary radioactive purity in 50 tons of solution, the project was changed to gallium metal. Using Davis’s idea, the extraction of minute quantities of $^{71}\text{Ge}$ from many tons of metallic gallium was independently developed. One advantage of metallic Ga is that it is significantly less sensitive to radioactive impurities. In 1980 an installation was built that contained 300 kg of Ga metal. In addition to testing the technology, this work also yielded a new limit on the law of conservation of electric charge [11]. By 1985 a pilot installation containing 7.5 tons of metallic gallium had been constructed at Troitsk.

The Soviet group built their experiment at the Baksan Neutrino Observatory in the Caucasus mountains. The first Ga exposure began in December 1989 and data collection has continued since that time. Gallex built their experiment at the Gran Sasso tunnel in Italy and collected data from 1991-1997. In 1998 they were reconstituted as the Gallium Neutrino Observatory (GNO) and they continued operation until 2003 [12].

3. SAGE

In 1986 the Soviet-American collaboration SAGE was officially established to carry out the gallium solar neutrino experiment at the Baksan Neutrino Observatory. The experiment is situated in a specially built deep underground laboratory where the measured muon flux is $(3.03 \pm 0.10) \times 10^{-9}/(\text{cm}^2 \text{s})$. It is located 3.5 km from the entrance of a horizontal adit excavated into the side of a mountain. The rock gives an overhead shielding equivalent to 4700 m of water and reduces the muon flux by a factor of $10^7$.

The mass of gallium used in SAGE at the present time is about 50 tonnes. It is in the form of liquid metal and is contained in 7 chemical reactors. A measurement of the solar neutrino capture rate begins by adding to the gallium a stable Ge carrier. The carrier is a Ga-Ge alloy with a known Ge content of approximately 350 $\mu$g and is distributed equally among all reactors. The reactor contents are stirred thoroughly to disperse the Ge throughout the Ga mass. After a typical exposure interval of four weeks, the Ge carrier and $^{71}\text{Ge}$ atoms produced by solar neutrinos and background sources are chemically extracted from the Ga using procedures described in [13, 14]. The final step of the chemical procedure is the synthesis of germane (GeH$_4$), which is used as the proportional counter fill gas with an admixture of (90–95)% Xe. The total efficiency of extraction is the ratio of mass of Ge in the germane to the mass of initial Ge carrier and is typically $(95 \pm 3)\%$. The systematic uncertainty in this efficiency is 3.4%, mainly arising from uncertainties in the mass of added and extracted carrier. The proportional counter is placed in the well of a NaI detector that is within a large passive shield and is counted for a typical period of 4–6 months.

Based on criteria described in [13], a group of events is selected from each extraction that are candidate $^{71}\text{Ge}$ decays. These events are fit to a maximum likelihood function [15], assuming that they originate from an unknown but constant-rate background and the exponentially decaying rate of $^{71}\text{Ge}$. A single run result has little significance because of its large statistical uncertainty.

The global best fit capture rate for all SAGE data from January 1990 through December 2005 (139
Figure 1. Results of all neutrino source experiments with Ga. The hashed region is the weighted average of the four experiments. See [20] for details.

runs and 264 separate counting sets) is \(66.2^{+3.5}_{-3.4}\) SNU, where the uncertainty is statistical only. If one considers the \(L\)-peak and \(K\)-peak data separately, the results are \(67.6^{+5.5}_{-5.3}\) SNU and \(65.5^{+4.7}_{-4.5}\) SNU, respectively. The agreement between the two peaks serves as a strong check on the robustness of the event selection criteria. The systematic effects fall into three main categories: those associated with extraction efficiency, with counting efficiency and with backgrounds. For a complete description of these effects see [13]. Including all uncertainties, our overall result is \(66.2^{+3.5}_{-3.4}\) (stat)\(^{+3.8}_{-3.4}\) (syst) SNU. If we combine the SAGE statistical and systematic uncertainties in quadrature, the result is \(66.2^{+5.2}_{-4.8}\) SNU.

The final result from 123 runs in the Gallex and GNO experiments is \(69.3 \pm 5.5\) (stat + syst) SNU [12]. The weighted combination of all the Ga experiments, SAGE, Gallex and GNO, is thus

\[
67.6 \pm 3.7 \text{ SNU. Present Ga experiment result.} \quad (1)
\]

It was very good that for many years there were two Ga experiments operating at the same time and it is indeed unfortunate that the GNO experiment was terminated for non-scientific reasons.

### 4. Source experiments

The experimental procedures of both Ga experiments, including the chemical extraction, counting and analysis techniques, have been checked by exposing the gallium target to reactor-produced neutrino sources whose activity was close to 1 MCi. Gallex has twice used \(^{51}\)Cr sources to irradiate their entire target; SAGE has irradiated about 25\% of their target with a \(^{51}\)Cr source and an \(^{37}\)Ar source [16, 17]. The results, expressed as the ratio \(R\) of the measured \(^{71}\)Ge production rate to that expected due to the source strength, are shown in Figure 4. The weighted average value of the ratio for the four experiments is \(R = 0.88 \pm 0.05\), more than two standard deviations less than unity.

Since other auxiliary tests, especially the \(^{74}\)As experiment of Gallex, have given great confidence in the knowledge of the various efficiencies in the Ga experiments, the combined result of these source tests should not be considered to be a measurement of the entire throughput of the Ga experiments. Rather, we believe that, although not statistically conclusive, the combination of these experiments suggests that the predicted rates may be overestimated. The most likely hypothesis \(^4\) is that the cross sections for neutrino capture to the lowest two excited states in \(^{71}\)Ge, both of which can be reached using either \(^{51}\)Cr or \(^{37}\)Ar sources, have been overestimated [19]. If the contribution of these two excited states to the predicted rate is set to zero, then \(R = 0.93 \pm 0.05\), reasonably consistent with unity. A new experiment with a considerably higher rate from the neutrino source is needed to settle this question.

As a side note, during the time of the SAGE \(^{37}\)Ar source experiment, which used 26 tonnes of Ga, solar neutrino extractions were also made from the remaining 22 tonnes of Ga. Since the SAGE counting

\(^4\) For an alternative explanation, based on transitions to sterile neutrinos, see [18].
system was filled with samples from the $^{37}$Ar source, we transported the $^{71}$Ge extracted from the solar runs to Gran Sasso, where GeH$_4$ was synthesized and the samples were counted in the GNO counting system. The combined result of six such solar runs was $64_{-22}^{+24}$ SNU [21], in excellent agreement with the overall result of the Ga experiments.

5. Comparison of gallium result to predictions of standard solar model
The capture rate $R_i$ of component $i$ of the solar neutrino spectrum is given in a radiochemical experiment by

$$R_i = \phi_i^\odot \langle P_i^{ee} \rangle \langle \sigma_i \rangle$$

where $\phi_i^\odot$ is the amplitude of the flux from this solar component at the production point in the Sun, $\langle P_i^{ee} \rangle$ is the integral over the solar spectrum of the probability of survival of the electron neutrino during its travel from where it is produced in the Sun to where it is detected at the Earth, and $\langle \sigma_i \rangle$ is the integral of the cross section for neutrino capture over the spectrum at the Earth. The physical origin for the reduction of the electron component of the solar neutrino flux is the now well-established mechanism of MSW neutrino oscillations [22].

Values of $\phi_i^\odot$, $\langle P_i^{ee} \rangle$ and $\langle \sigma_i \rangle$ are given for each neutrino component in Table 2. The fluxes are from two solar models with differing composition [23]. The other quantities were calculated assuming three-neutrino mixing to active neutrinos with parameters $\Delta m_{12}^2 = (7.92 \pm 0.36) \times 10^{-5}$ eV$^2$, $\theta_{12} = 34.1_{-1.5}^{+1.7}$ degrees and $\theta_{13} = 5.44_{-2.79}^{+2.79}$ degrees [24]. The approximate formulae given in [25] were used for the survival probability $P_i^{ee}(E)$. Since radiochemical experiments average over a long exposure interval, regeneration in the Earth was neglected. The cross sections $\sigma(E)$ were taken from [26] but were modified to delete the effect of the lowest two excited states in $^{71}$Ge according to the results of the neutrino source experiments as given in the previous section. The neutrino spectra were taken from [26] ($pp$ and CNO), [27] ($^8$B) and [28] ($hep$).

There is excellent agreement between the calculated ($67.3^{+3.9}_{-3.5}$ SNU) and observed ($67.6 \pm 3.7$ SNU) capture rates in $^{71}$Ga.

Table 2. Factors needed to compute the capture rate in $^{71}$Ga solar neutrino experiments. The units of flux are $10^{10}$ ($pp$), $10^9$ ($^7$Be), $10^8$ ($pep$, $^{13}$N,$^{15}$O), $10^6$ ($^8$B,$^{17}$F), and $10^5$ ($hep$) cm$^{-2}$s$^{-1}$. The uncertainty values are at 68% confidence.

| Spectrum | Flux $\phi_i^\odot$ | $\langle P_i^{ee} \rangle$ | $\langle \sigma_i \rangle$ | Capture rate $R_i$ (SNU) |
|----------|------------------|----------------|----------------|-----------------|
|          | BP04             | BP04+         | (10$^{-46}$ cm$^2$) | BP04            | BP04+ |
| $pp$     | 5.94(1$^{+0.01}_{-0.01}$) | 5.99 | 0.555(1$^{+0.038}_{-0.040}$) | 11.75(1$^{+0.024}_{-0.023}$) | 38.7(1$^{+0.046}_{-0.047}$) | 39.1 |
| $pep$    | 1.40(1$^{+0.02}_{-0.02}$)  | 1.42 | 0.517(1$^{+0.033}_{-0.034}$) | 194.4(1$^{+0.17}_{-0.024}$) | 1.41(1$^{+0.17}_{-0.046}$) | 1.43 |
| $^7$Be    | 4.86(1$^{+0.12}_{-0.12}$)  | 4.65 | 0.537(1$^{+0.036}_{-0.037}$) | 68.22(1$^{+0.070}_{-0.023}$) | 17.8(1$^{+0.14}_{-0.13}$) | 17.0 |
| $^{13}$N  | 5.71(1$^{+0.37}_{-0.35}$)  | 4.06 | 0.539(1$^{+0.038}_{-0.038}$) | 56.86(1$^{+0.099}_{-0.023}$) | 1.75(1$^{+0.38}_{-0.35}$) | 1.24 |
| $^{15}$O  | 5.03(1$^{+0.43}_{-0.39}$)  | 3.54 | 0.531(1$^{+0.035}_{-0.036}$) | 107.2(1$^{+0.13}_{-0.023}$) | 2.86(1$^{+0.45}_{-0.39}$) | 2.02 |
| $^{17}$F  | 5.91(1$^{+0.44}_{-0.44}$)  | 3.97 | 0.531(1$^{+0.035}_{-0.036}$) | 107.8(1$^{+0.13}_{-0.023}$) | 0.03(1$^{+0.46}_{-0.44}$) | 0.02 |
| $^8$B     | 5.79(1$^{+0.23}_{-0.23}$)  | 5.26 | 0.374(1$^{+0.044}_{-0.039}$) | 21580(1$^{+0.32}_{-0.15}$) | 4.67(1$^{+0.40}_{-0.28}$) | 4.25 |
| $hep$    | 7.88(1$^{+0.16}_{-0.16}$)  | 8.04 | 0.347(1$^{+0.061}_{-0.054}$) | 66300(1$^{+0.33}_{-0.16}$) | 0.02(1$^{+0.37}_{-0.23}$) | 0.02 |
| **Total** |               |             |                   | 67.3$^{+3.9}_{-3.5}$ | 65.1 |
6. The $pp$ neutrino flux

One of the main purposes of the Ga experiment is to provide information that leads directly to the experimental determination of the flux of $pp$ neutrinos at the Earth. In this Section we will assume the Sun is generating energy by the $pp$ cycle, and not dominantly by the CNO cycle, and will derive the present best value for the $pp$ flux directly from the results of neutrino experiments.

To obtain the $pp$ flux we begin with the combined capture rate from the SAGE and GALLEX/GNO experiments given above of 67.6±3.7 SNU. This rate is the sum of the rates from all the components of the solar neutrino flux, which we denote by $[\phi_{pp}^7\text{Be}+\text{CNO}+\text{pep}^8\text{B}]\text{Ga}$. (We ignore the hep contribution.)

The only one of these flux components that is known from direct experiment is the $^8\text{B}$ flux, measured by SNO to be $[^8\text{B}]\text{SNO} = (1.68 \pm 0.11) \times 10^6$ electron neutrinos/(cm$^2$-s) [29] at the Earth. We multiply this flux by the cross section for $^8\text{B}$ given in Table 2 and find that the contribution to the Ga experiment is $[^8\text{B}]\text{Ga} = 3.7^{+1.2}_{-0.7}$ SNU. Subtracting this measured value from the total Ga rate gives $[\phi_{pp}^7\text{Be}+\text{CNO}+\text{pep}^8\text{B}]\text{Ga} = 64.0^{+3.7}_{-3.9}$ SNU.

The measured capture rate in the Cl experiment is $[^7\text{Be}+\text{CNO}+\text{pep}^8\text{B}]\text{Cl} = 2.56 \pm 0.23$ SNU [4]. In a manner analogous to Ga we can calculate the cross section for $^8\text{B}$ neutrinos on $^{37}\text{Cl}$, including the suppression factor, to be $1.02(1 \pm 0.046) \times 10^{-42}$ cm$^2$. We multiply this by the flux measured by SNO and deduce that the contribution of $^8\text{B}$ to the Cl experiment is $[^8\text{B}]\text{Cl} = 1.72 \pm 0.14$ SNU. Subtracting this component from the total leaves $[^7\text{Be}+\text{CNO}+\text{pep}]\text{Cl} = 0.84 \pm 0.27$ SNU, all of which is due to neutrinos of medium energy.

We assume the Sun is generating its energy via the $pp$ cycle so these medium-energy neutrinos are dominated by $^7\text{Be}$. We can thus make the approximation that $[^7\text{Be}+\text{CNO}+\text{pep}]\text{Cl} \times$ cross section for $^7\text{Be}$ on Ga/cross section for $^7\text{Be}$ on Cl = $(0.84 \pm 0.27) / [23.9 (1 + 0.07) / [2.40 (1 \pm 0.02)]] = 23.9^{+7.9}_{-7.6}$ SNU. There is an additional error due to the approximation used, which is estimated to be 10%, giving the result $[^7\text{Be}+\text{CNO}+\text{pep}]\text{Ga} = 23.9^{+8.1}_{-8.0}$ SNU.

We subtract this contribution from the rate given above and get the result for the measured $pp$ rate in the Ga experiment $[\phi_{pp}\text{Ga}] = [\phi_{pp}^7\text{Be}+\text{CNO}+\text{pep}]\text{Ga} - [^7\text{Be}+\text{CNO}+\text{pep}]\text{Ga} = 40.1^{+6.6}_{-6.9}$ SNU. Dividing this capture rate by the cross section for capture of $pp$ neutrinos of $11.8(1^{+0.024}_{-0.024}) \times 10^{-46}$ cm$^2$ gives the measured electron neutrino $pp$ flux at Earth of $\phi_{pp}^5 = (3.41^{+0.76}_{-0.77}) \times 10^{10}$/(cm$^2$-s). The major component of the error in this $pp$ flux measurement is due to the poor knowledge of the medium-energy neutrinos which was inferred from the Cl experiment.

For comparison, the standard solar model calculates the $pp$ flux produced in the Sun to be $\phi_{pp}^\odot = 5.94(1 \pm 0.01) \times 10^{10}$/(cm$^2$-s) [23] $^5$. If we multiply this rate by the average survival probability for $pp$ neutrinos, which from Table 2 is 0.555(1$^{+0.038}_{-0.040}$), we obtain a $pp$ flux at the Earth of $\phi_{pp}^5 = (3.30^{+0.15}_{-0.14}) \times 10^{10}$/(cm$^2$-s), in excellent agreement with the value determined above from solar neutrino experiments.

In the future it will be possible to reduce the error in this flux measurement when there are new experiments that directly measure the $^7\text{Be}$ flux, as anticipated by Borexino and KamLAND, and the CNO flux, as anticipated by SNO+. The dominant error should eventually be due to the inaccuracy of the Ga measurement itself.

7. Is the neutrino capture rate in Ga constant?

Short-term variations in the Gallex-GNO rate with periods from 15 days to a few 100 days have been considered by Pandola [30] and Sturrock et al. [31]. Pandola’s analysis finds no variability but Sturrock et al. see evidence for variation if one considers the Gallex and GNO data sets separately.

The possibility of variability over longer time periods has been considered by several authors [32, 33]. In a plot of the data there appears to be a difference between early and late time periods, which gives a visual hint of a long-term decrease, as illustrated in Figure 2. The Gallex-GNO data is shown on the left

$^5$ The error here is only 1% because the measured solar luminosity was used in this calculation.
of this Figure where the data have been grouped by the experimenters into 7 intervals. The SAGE data, divided into intervals of one calendar year, is shown on the right of Figure 2. The average rate prior to 1997 is higher in both experiments than in the data after 1997.

If one assumes the rate in Gallex-GNO varies linearly in time then the best fit gives [12]

\[
\text{Capture rate} = 82 \pm 10 - (1.7 \pm 1.1) \times [t\text{(year)} - 1990].
\]  

(3)

These trend lines are plotted for both experiments in Figure 2 and there is reasonably good visual agreement with the measured data.

When examined quantitatively, however, the evidence for long-term variability becomes less convincing. A \( \chi^2 \) test applied to the Gallex-GNO data with (without) the assumed time variation yields \( \chi^2/\text{dof} = 10.8/5 \) (13.2/6), prob. = 5.6\% (4.0\%), i.e., the fit to both the time-varying rate and to a constant rate is more or less equally bad. For the SAGE data the fit to a constant rate gives \( \chi^2/\text{dof} = 11.7/16 \), prob. = 76\%, whereas the fit to the central Gallex-GNO trend line yields \( \chi^2/\text{dof} = 11.4/17 \), prob. = 83\%, i.e., the fit to both rate hypotheses is quite good. At the present time we cannot differentiate between these two hypotheses, but it should become possible to do so with considerable additional data.

Up to now it is not known if this apparent variability is a statistical fluctuation or an indication of a real effect, such as has been considered by Pulido et al. [34].

8. Other radiochemical experiments

Several other radiochemical experiments to measure solar neutrinos have been developed to various degrees. These include \( ^{127}\text{I} \rightarrow ^{127}\text{Xe} \) [35, 36] and \( ^{81}\text{Br} \rightarrow ^{81}\text{Kr} \) [37] experiments that would in many ways be similar to the \( ^{37}\text{Cl} \) experiment, and a \( ^{97}\text{Mo} \rightarrow ^{97}\text{Tc} \) [38] experiment that could measure the long-term history of the \( ^{8}\text{B} \) solar neutrino flux. Although very considerable efforts were expended in the United States on the \( ^{127}\text{I} \) and \( ^{97}\text{Mo} \) experiments, they were never brought to fruition, mainly because of a lack of funding.

The \( ^{7}\text{Li} \rightarrow ^{7}\text{Be} \) experiment has continued to be pursued in Russia. Methods for the efficient extraction of Be from metallic Li have been proven [39] and an experiment could, in principle, be built [40].

At the present time interest in radiochemical experiments has greatly decreased and it is only direct-counting experiments that are under development. Nonetheless, the radiochemical experiments stimulated great interest in the solar neutrino problem, which led to the real-time experiments Super-Kamiokande, SNO and KamLAND.
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