

³⁷Ar as a calibration source for solar neutrino detectors

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I discuss the possibility that a high-intensity 811-keV ³⁷Ar neutrino source, produced by neutron capture on separated ³⁶Ar, could be used to calibrate the ⁷Be solar neutrino capture cross sections of ⁷¹Ga, ¹²⁷I, and other detectors.

Calibration tests of solar neutrino detectors are important both to verify detector efficiencies and to eliminate uncertainties in nuclear cross sections.¹ Several calibration sources have been discussed previously²⁻⁴

$$\begin{aligned}
 e^- + {}^{65}\text{Zn} &\rightarrow \begin{cases} {}^{65}\text{Cu} + \nu(1.343 \text{ MeV}, 47.8\%), \\ {}^{65}\text{Cu}^* + \nu(0.227 \text{ MeV}, 50.75\%), \end{cases} \quad \tau_{1/2} = 244.1 \text{ d}, \\
 e^- + {}^{51}\text{Cr} &\rightarrow \begin{cases} {}^{51}\text{V} + \nu(0.746 \text{ MeV}, 90.1\%), \\ {}^{51}\text{V}^* + \nu(0.426 \text{ MeV}, 9.9\%), \end{cases} \quad \tau_{1/2} = 27.7 \text{ d}, \\
 e^- + {}^{152}\text{Eu} &\rightarrow \begin{cases} {}^{152}\text{Sm}^* + \nu(1.048 \text{ MeV}, 1.3\%), \\ {}^{152}\text{Sm}^* + \nu(0.772 \text{ MeV}, 22\%), \\ {}^{152}\text{Sm}^* + \nu(0.624 \text{ MeV}, 17\%), \\ {}^{152}\text{Sm}^* + \nu(0.328 \text{ MeV}, 25\%), \\ {}^{152}\text{Sm}^* + \nu(0.278 \text{ MeV}, 2.2\%), \end{cases} \quad \tau_{1/2} = 13 \text{ yr}.
 \end{aligned}$$

The discrete energies of the emitted neutrinos and the corresponding branching ratios are given in parentheses. In particular, the Gallex collaboration plans to use an intense (~1 MCi) ⁵¹Cr source to calibrate the ⁷¹Ga experiment:⁵ the 746-keV neutrino will excite all three Gamow-Teller transitions [to the $\frac{1}{2}^-$ ground ($E_{ex} = 236 \text{ keV}$), $\frac{5}{2}^-$ ($E_{ex} = 411 \text{ keV}$), and $\frac{3}{2}^-$ ($E_{ex} = 736 \text{ keV}$) states in ⁷¹Ge] that contribute to the capture of ⁷Be solar neutrinos ($E_\nu = 862 \text{ keV}$).

The original motivation for the work reported here was the observation that none of the above sources could provide an unambiguous calibration of the ⁷Be neutrino cross section for the proposed ¹²⁷I detector.⁶ The transition excited by ⁷Be neutrinos, to the $\frac{3}{2}^+$ state lying at 125 keV in ¹²⁷Xe, has an excitation threshold of 789 keV. Thus the ⁵¹Cr neutrinos and the principle ¹⁵²Eu neutrinos are too low in energy. The 1.343-MeV neutrinos from ⁶⁵Zn are too energetic, exciting several additional Gamow-Teller (GT) transitions.⁷ While the ¹²⁷I experiment is very attractive technically and potentially inexpensive, uncertainties in estimating the ⁷Be and ⁸B neutrino absorption cross sections are a concern. Thus, a calibration of the transition to the $\frac{3}{2}^+$ level is very important: in addition to determining the ⁷Be cross section, it could provide a normalization for (p, n) mappings of the GT strength. The normalization is a primary uncertainty in (p, n) calibrations of ⁸B neutrino cross sections.⁸

A search of the table of isotopes revealed one practical calibration source for the ¹²⁷I experiment, ³⁷Ar ($\tau_{1/2} = 35 \text{ d}$). Several properties of this nucleus are also very attractive from the perspective of a ⁷¹Ga calibration, so the dis-

ussion below will concentrate on the general attributes of ³⁷Ar as a neutrino source.

Neutrino yield. ³⁷Ar decays exclusively to the ground state of ³⁷Cl, generating a monoenergetic neutrino with $E_\nu = 811 \text{ keV}$. (More correctly, this is the K -capture neutrino energy. L capture occurs with a 10% probability, yielding a neutrino with $E_\nu = 814 \text{ keV}$.) Thus, in contrast to ⁶⁵Zn, ⁵¹Cr, and ¹⁵²Eu, no energetic nuclear γ rays are produced, greatly lessening safety concerns, as discussed below. Because of the higher neutrino energy and the 100% branching ratio to the ³⁷Cl ground state, the cross section for exciting nuclear targets exceeds that for ⁵¹Cr. Comparative results for the relevant states in ⁷¹Ge (Table I) show that the ³⁷Ar neutrino cross section is 20% to 44% larger. In addition, the longer half-life of ³⁷Ar provides 26% more neutrinos. The net result, in the case of ⁷¹Ge, is that a 1-MCi ³⁷Ar source is equivalent to 1.5 MCi (1.8 MCi) of ⁵¹Cr for exciting the ground state (500 keV state) in ⁷¹Ge.

TABLE I. Neutrino cross sections for exciting states in ⁷¹Ge and ¹²⁷Xe by ⁵¹Cr, ³⁷Ar, and ⁷Be neutrinos, taking $|M^2| = 1$ for all transitions. The units are 10^{-44} cm^2 .

Source	⁷¹ Ge (g.s.)	⁷¹ Ge (175 keV)	⁷¹ Ge (500 keV)	¹²⁷ Xe (125 keV)
⁵¹ Cr	3.88	2.59	0.85	0.0
³⁷ Ar	4.66	3.24	1.22	3.25
⁷ Be	4.75	3.25	1.33	3.41

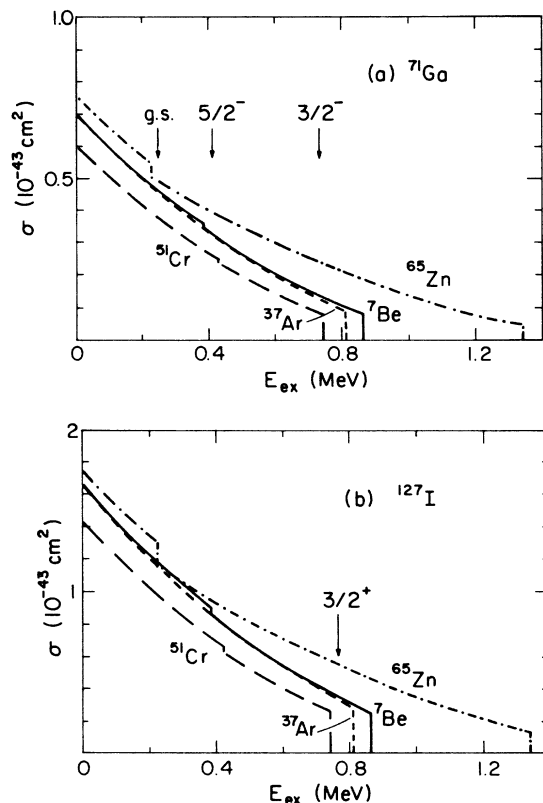


FIG. 1. Neutrino cross sections in (a) ⁷¹Ga and (b) ¹²⁷I as functions of the atomic excitation energy for various neutrino sources, assuming a fixed nuclear matrix element $|M^2| = 1$.

In Fig. 1, cross sections for ⁷Be, ³⁷Ar, ⁵¹Cr, and ⁶⁵Zn neutrinos, assuming a fixed nuclear matrix element $|M^2| = 1$, are shown as a function of the atomic excitation energy E_{ex} . Note that the ³⁷Ar neutrino cross section tracks that for ⁷Be solar neutrinos remarkably well, both absolutely and relatively. The latter is important, when, as in the case of ⁷¹Ga, more than one nuclear transition contributes. The ³⁷Ar/⁷Be and ⁵¹Cr/⁷Be cross section ratios are 0.96 ± 0.04 and 0.73 ± 0.09 over the ⁷Be neutrino excitation range for ⁷¹Ga ($236 \text{ keV} \leq E_{ex} \leq 736 \text{ keV}$). Thus, an ³⁷Ar source mimics the ⁷Be solar neutrino phase space a factor of three more accurately than does a ⁵¹Cr source.

Production of ³⁶Ar. ³⁷Ar can be produced by irradiating enriched ³⁶Ar (natural abundance 0.337%) in the intense neutron flux of a reactor. The relevant data on Ar isotopes are given in Table II. The ³⁶Ar(*n*, γ) cross section of $\sigma = 5.2 \pm 0.5 \text{ b}$ is almost an order of magnitude

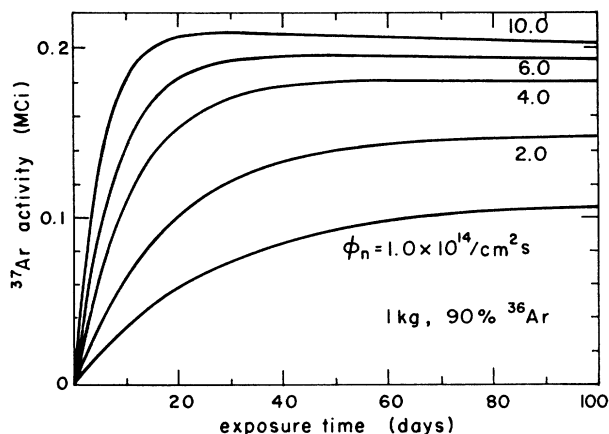


FIG. 2. The ³⁷Ar activity as a function of irradiation time for various values of the neutron flux ϕ_n .

larger than that for ⁴⁰Ar, the principal stable isotope. Thus, any enrichment above 10% makes ³⁶Ar the primary neutron absorber.

The very large (*n*, α) and (*n*,*p*) cross sections for ³⁷Ar, 1970 and 69 b, lead to unfortunate “burn-up” problems⁹ for the product isotope, limiting the achievable ³⁷Ar/³⁶Ar ratio to no more than 0.0025. This limit is equivalent to a maximum activity of 0.23 MCi for a 1-kg target, assuming a ³⁶Ar content of 90%. The most straightforward solution to this problem, irradiating 5–10 kg of ³⁶Ar to produce a ~1-MCi source, appears to be practical, as discussed below. However, if the burn-up problem could be overcome, a 1-MCi source could be generated from as little as 0.6 kg of ³⁶Ar (assuming a neutron intensity of $\phi_n \sim 10^{15}/\text{cm}^2\text{s}$ for the resulting compact target). Thus, it might be profitable to explore recirculation systems for removing ³⁷Ar during irradiation, or the feasibility of extracting ³⁷Ar between a series of short, intense irradiations. Alternatively, one might explore whether the ³⁶Ar(*n*, γ)/³⁷Ar(*n*, γ) ratio is more favorable at non-thermal energies, such as those available at “fast flux” reactors. Although ³⁷Ar can also be produced by ⁴⁰Ca(*n*, α), the thermal neutron cross section is very small ($2.5 \pm 1.1 \text{ mb}$) because of the unfavorable *Q* value (1.75 MeV). Thus, this option would require an intense, energetic neutron source.

The most likely strategy is to use a larger ³⁶Ar target to compensate for the burn-up problem. In Fig. 2, the ³⁷Ar activity is plotted as a function of exposure time for a 1-kg target containing 90% ³⁶Ar, 5% ³⁸Ar, and 5% ⁴⁰Ar. The activity calculations coupled all the isotopes in Table I, the ³⁷Ar(*n*,*p*) and (*n*, α) products ³⁷Cl and ³⁴S, and their

TABLE II. Abundances, half-lives, and thermal neutron capture cross sections (Ref. 14) for argon isotopes.

	³⁶ Ar	³⁷ Ar	³⁸ Ar	³⁹ Ar	⁴⁰ Ar	⁴¹ Ar	⁴² Ar
Abundance	0.337	...	0.063	...	99.60
σ_n^* (b)	5.2 ± 0.5	2040 ± 340	0.8 ± 0.2	600 ± 300	0.66 ± 0.01	0.5 ± 0.1	?
$\tau_{1/2}$...	35 d	...	269 yr	...	1.83 h	33 yr

*These are (*n*, γ) cross sections except for ³⁷Ar, where $\sigma_n \approx \sigma(n,p) + \sigma(n,\alpha) = (69 \pm 14 \text{ b}) + (1970 \pm 330 \text{ b})$.

(n, γ) daughters ^{38}Cl and ^{35}S . For a 40-d exposure, an activity of 0.178 MCi is achieved with a neutron flux of $4 \times 10^{14}/\text{cm}^2\text{s}$. As this activity is 75% of the theoretical maximum, higher fluxes, even if attainable, would not be of significant benefit. Reducing ϕ_n by a factor of four (to $10^{14}/\text{cm}^2\text{s}$) halves the activity (0.086 MCi). For definiteness, in the calculations below I assume $\phi_n = 2 \times 10^{14}/\text{cm}^2\text{s}$, yielding an activity of 0.134 MCi. Thus, 7.5 kg of target (90% ^{36}Ar) would yield a 1-MCi source after 40 d of irradiation.

At 0°C and one atmosphere of pressure the density of argon is 1.78 g/l. At -189.4°C argon liquefies, with the fluid density being 1.40 g/l. This places a lower bound on the dimensions of the source. At liquid density the neutron absorption length in enriched argon (90% ^{36}Ar) is 9.1 cm, a value that will drop to 5.8 cm at the end of the 40-d irradiation described above. To keep the target relatively transparent to neutrons, a maximum thickness of about 3 cm is then appropriate. Thus, the minimum dimensions of a 7.5-kg target are $3 \text{ cm} \times (40 \text{ cm})^2$. It may prove possible to irradiate a liquid target, despite the engineering problems this presents. Alternatively, a highly compressed gas could be used both in the irradiation and calibration phases. Since densities approaching those of a liquid are achievable, no large increase in target dimensions should result.

An argon source is attractive because gas diffusion, distillation, and other technologies exist for separating ^{36}Ar . For instance the 213-m cryogenic distillation column operated by Los Alamos National Laboratory can produce about 12 kg of ^{36}Ar in 2.5 yr of operation.¹⁰ Production costs are modest, and enrichments in excess of 90% can likely be achieved (e.g., 92% ^{36}Ar , 4% ^{38}Ar , and 4% ^{40}Ar).¹⁰ The production of such quantities of ^{36}Ar could be of value to the ^{71}Ga , ^{127}I , and other future experiments^{11,12} (e.g., ^{81}Br , ^{115}In): as very little of the ^{36}Ar is depleted by the irradiation, sources could be recycled.

Safety aspects. The absence of a nuclear gamma ray accompanying the decay of ^{37}Ar is important from the standpoint of safety. While the 320-keV γ emitted in ^{51}Cr decay can be fully shielded by about 2 cm of lead,¹³ the possibility of a rupture in the containment vessel must be a constant concern in transporting the source from the reactor to the underground laboratory, and in handling at the laboratory. The ^{37}Ar decay is accompanied only by x rays emitted in atomic rearrangement: the K-electron binding energy is 3.21 keV.

The principal concern among other isotopes of argon is the production of ^{41}Ar ($\tau_{1/2} = 1.83 \text{ h}$) from ^{40}Ar . The activity in ^{41}Ar will be maintained at $\sim 22 \text{ kCi}$ during irradiation, assuming that $\phi_n = 2 \times 10^{14}/\text{cm}^2\text{s}$ and that the 7.5-kg target contains 5% ^{40}Ar . (The ^{41}Ar activity varies as the product of the ^{40}Ar content and ϕ_n .) The dominant ^{41}Ar β^- decay branch (99.2%) emits electrons with kinetic energies up to 1.2 MeV. With the short half-life, this activity can be allowed to decay away before the source is moved from the reactor site. If the target contains 5% ^{38}Ar , the activity in long-lived ^{39}Ar ($t_{1/2} = 269 \text{ yr}$) is much lower (6.2 Ci). The β^- decay of ^{39}Ar produces

electrons with energies up to 565 keV.

The corresponding activity from ^{38}Cl ($\tau_{1/2} = 37.3 \text{ min}$), produced by $^{37}\text{Ar}(n,p)^{37}\text{Cl}$ followed by $^{37}\text{Cl}(n,\gamma)^{38}\text{Cl}$ ($\sigma_{n\gamma} = 0.43 \text{ b}$), is $\sim 210 \text{ Ci}$ after the 40-d irradiation. The decay to the ground state of ^{38}Ar is strong (58% branch) and produces high-energy electrons ($\lesssim 4.92 \text{ MeV}$). It is again fortunate that this activity is short-lived. The activity from ^{35}S ($\tau_{1/2} = 87.4 \text{ d}$), produced by $^{37}\text{Ar}(n,\alpha)^{34}\text{S}$ followed by $^{34}\text{S}(n,\alpha)^{35}\text{S}$ ($\sigma_{n\gamma} = 0.23 \text{ b}$), is low (20 Ci) and produces only low-energy electrons ($\lesssim 168 \text{ keV}$).

Because the target is a gas, its purity should be unusually high, minimizing concerns about high-energy γ 's emitted from irradiated trace elements. Radioactivity from the vessel containing the argon would have to be studied.

Summary. It appears that ^{37}Ar could provide an unusually safe and intense neutrino source. The economics may be quite favorable because highly enriched ^{36}Ar can be produced in multikilogram quantities at existing facilities. As the ^{37}Ar neutrinos mimic the ^7Be solar neutrino phase space remarkably well, this source could be used to measure the ^7Be neutrino cross sections for targets like ^{71}Ga , ^{81}Br , and ^{115}In . In the case of ^{127}I , where the ^7Be neutrino transition has a high threshold, ^{37}Ar appears to be unique as a potential calibration source. Thus, from a theoretical viewpoint, a ^{36}Ar source appears to have some merit. On the other hand, a more serious investigation must address many other concerns. To what densities can the target be compressed? What is the resultant irradiation strategy and maximum ϕ_n ? How thick must the containment vessel be and what effect will it have on ϕ_n ? These and other engineering questions are presently unanswered.

Note added in proof. Two comments made recently to me are quite intriguing. William Wilkes suggests that an on-line single-pass extraction system could be built to remove ^{37}Ar from the ^{36}Ar source during irradiation, thereby avoiding the burn-up problem. One would then need only $\sim 1 \text{ kg}$ of ^{36}Ar to produce a 1-MCi source. Ray Davis points out that argon can be flushed from various calcium compounds by a helium purge. Thus ^{37}Ar could be milked from a large ^{40}Ca target exposed in some energetic neutron flux. The $^{40}\text{Ca}(n,\alpha)$ cross section at higher neutron energies is known¹⁵ ($\sigma_n \sim 59 \text{ mb}$ at 3 MeV and $\gtrsim 200 \text{ mb}$ at 5.7–7.5 MeV). A modest cross section could be quite satisfactory because the target material is inexpensive. Each of these schemes has the advantage of producing an extremely compact source that might be of interest in various terrestrial neutrino experiments. For instance, Wilkes's proposal might yield a 100-g 1-MCi source ($\sim 10\%$ ^{37}Ar).

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