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## On atmospheric $^{39}\text{Ar}$ and $^{42}\text{Ar}$ abundance

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Received 10 September 1994

### Abstract

$^{39}\text{Ar}$  and  $^{42}\text{Ar}$  are possible sources of background in large volume argon based detectors for the study of low energy neutrino events, as proposed for the ICARUS experiment. The production of these nuclides from neutron capture by atmospheric argon is estimated by taking into account both cosmic rays and contributions from nuclear explosions.

### 1. Introduction

The presence of  $^{39}\text{Ar}$  and  $^{42}\text{Ar}$  in commercially available argon has often been assumed. These nuclides have very long half lives, 269 and 33 yr respectively, and, considering their decay energies and decay modes ( $\beta$  particles up to 565 keV for  $^{39}\text{Ar}$  and up to 3520 keV for  $^{42}\text{Ar}$ ), can be important sources of background in a large volume argon based detector for the study of low energy neutrino events.

Both nuclides can be produced in spallation reactions, electromagnetic interactions with cosmic muons [1], or by neutron interactions with stable argon isotopes ( $(n, \gamma)$  or  $(n, 2n)$ ). None of the nuclides present in an appreciable amount in the atmosphere can be a target for argon production by the first mechanism.  $^{39}\text{Ar}$  can be produced by the second mechanism or by neutron capture on  $^{38}\text{Ar}$  or by the  $(n, 2n)$  reaction on  $^{40}\text{Ar}$ , while the only reliable production

mode for  $^{42}\text{Ar}$  appears to be double neutron capture by  $^{40}\text{Ar}$ .

The ICARUS experiment [2] at the Gran Sasso Laboratory, now in the final design phase, is proposing, among other physics items, to study solar neutrinos. ICARUS can detect solar neutrinos from the  $^8\text{B}$  part of the cycle by observing electrons produced in the following two reactions

$$(a) \nu_{e,\mu,\tau} + e^- \rightarrow \nu_{e,\mu,\tau} + e^- \quad (\text{elastic scattering}),$$

$$(b) \nu_e + {}^{40}\text{Ar} \rightarrow {}^{40}\text{K}^* + e^- \quad (\text{absorption on Ar nucleus}).$$

Reaction (a) is characterized by a single electron up to 15 MeV, while reaction (b) produces one electron (up to  $\sim 9$  MeV) accompanied by several photons. Radioactive decays from argon isotopes inside the liquid could be sources of background in the lower part of the electron energy spectrum.  $^{42}\text{Ar}$  is by far the most troublesome from this point of view.

A very crude estimate of an upper limit on the  $^{42}\text{Ar}$  concentration in natural liquid argon (LAR) can be derived from the total counting rate measurement performed with

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the ICARUS 3-ton prototype [4], running at the ground level at CERN.

Monitoring the number of contained events that give an energy deposit between 2 and 4 MeV in a sensitive volume of 18 l of LAr, we observed a rate of 0.77 Hz.

This rate can be only partially attributed to the presence of  $^{42}\text{Ar}$  because it includes a large contribution from the radioactivity of the dewar and of many nearby concrete blocks used in the experimental set-up. Moreover, a contribution from cosmic rays surviving the cuts from the external trigger system has also to be taken into account.

An upper limit on the  $^{42}\text{Ar}$  concentration could be obtained from this measurement considering the total rate as entirely due to  $^{42}\text{Ar}$  presence. With this assumption, a limit of the order of  $4 \times 10^{-18}$   $^{42}\text{Ar}$  atom/ $^{40}\text{Ar}$  atom can be inferred.

Recently a dedicated search for  $^{42}\text{Ar}$  in natural liquid argon was performed at the Gran Sasso Laboratory by a collaboration between the University of Milan, the ICARUS group and the LNGS by means of gamma spectroscopy with a germanium detector [3a]. The resulting upper limit on the  $^{42}\text{Ar}$  concentration in natural liquid argon is  $1.2 \times 10^{-18}$   $^{42}\text{Ar}$  atoms/ $^{40}\text{Ar}$  atom at 90% C.L. This is consistent with the other known result [3b]. However, the measurement has been accidentally affected by an abnormal high concentration of radon, due to a failure of the ventilation system in the experimental site.

This upper limit is equivalent to a specific activity of  $1.8 \times 10^{-2}$  Bq/l-of-LAr, corresponding to a total rate of about  $6 \times 10^4$  Hz in the 4.7 kton liquid argon ICARUS module. The rate in the higher part of the energy spectrum, for example between 3 and 3.5 MeV, could be as high as 200 Hz. In our 3 ton prototype we achieved an energy resolution of 7% [4] in this energy range. Supposing a similar resolution in the 4.7 kton module, a 5 MeV threshold is at  $7\sigma$  from the 3 to 3.5 MeV bin. If we assume a Gaussian energy resolution function we can estimate that the number of expected events over threshold should not exceed a few tens of events per year, to be compared to about 4200 elastic scattering events and 3800 absorption events per year, as foreseen from the Standard Solar Model calculations [2]. We can conclude that under these assumptions,  $^{42}\text{Ar}$  is not a serious background. However, a more stringent upper limit on the  $^{42}\text{Ar}$  concentration would permit us to take into account a possibly worse energy resolution, the non-Gaussian behaviour of the tails in the energy resolution function and the possibility to lower the energy threshold.

In what follows we derive a new estimate of the upper limit of the  $^{42}\text{Ar}$  concentration in natural argon. We will consider  $^{39}\text{Ar}$  and  $^{42}\text{Ar}$  production according to the following schemes

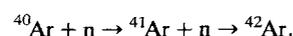
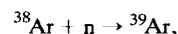


Table 1  
Parameters used in computations [7]

| Nuclide          | % Abundance | Reaction       | Cross section<br>(b) $\sigma_0$ | Half-life |
|------------------|-------------|----------------|---------------------------------|-----------|
| $^{38}\text{Ar}$ | 0.063       | (n, $\gamma$ ) | 0.8                             | –         |
| $^{39}\text{Ar}$ | –           | (n, $\gamma$ ) | 600                             | 269 yr    |
| $^{40}\text{Ar}$ | 99.6        | (n, $\gamma$ ) | 0.64                            | –         |
| $^{41}\text{Ar}$ | –           | (n, $\gamma$ ) | 0.5                             | 1.827 h   |
| $^{42}\text{Ar}$ | –           | –              | –                               | 32.9 yr   |
| $^{14}\text{N}$  | 99.63       | (n, p)         | 1.82                            | –         |
| $^{14}\text{C}$  | –           | (n, $\gamma$ ) | $< 10^{-6}$                     | 5730 yr   |

Computations are based on the nuclear data compiled in Table 1 [7], on an estimate of the naturally occurring atmospheric neutron flux, and on some considerations about nuclear explosion effects.

## 2. Naturally occurring neutrons — steady state conditions

The cross-sections for the reactions of interest are poorly known, in particular, for what concerns their dependence on neutron energy. For the moment we will assume a cross-section proportional to  $1/\nu$ , the inverse of the neutron velocity. In this case the reaction rate per atom is given by

$$R = \sigma_0 n v_0 = \sigma_0 \phi, \quad (1)$$

where

$$n = \int_0^\infty n'(v) dv \quad (2)$$

is the total neutron density ( $n/\text{cm}^3$ ), i.e. a quantity independent of the neutron energy spectrum,  $\sigma_0$  is the cross section at a reference velocity ( $v_0$ ) (for thermal neutrons  $v_0 = 2200$  m/s) and  $\phi = n v_0$  is the conventional neutron flux. Since the atmosphere is an infinite and poorly absorbing medium, thermal neutrons are likely to be the main component of the neutron flux. This last quantity ( $\phi$ ) has been measured several times, but the reported values are largely inconsistent (see e.g. Refs. [5,6]); moreover it is known to vary with place and time. We have derived an estimate of the mean atmospheric neutron flux from  $^{14}\text{C}$  (produced by the well known reaction  $^{14}\text{N}(n, p)^{14}\text{C}$ ) specific activity whose value is known with reasonable accuracy. Disregarding a number of corrections of minor importance in our case, a good mean equilibrium value for the  $^{14}\text{C}$  specific activity, before the large scale fossil fuel burning and nuclear experiments in the atmosphere, is 0.25 decay per second (dps)/g-of-carbon [5]. The mean value in the atmosphere of the steady state  $^{14}\text{C}$  specific activity ( $I(14)$ ) is

$$I(14) = N(14)\bar{R}, \quad (3)$$

where  $N(14)$  is the concentration of nitrogen atoms and  $\bar{R}$  is the mean reaction rate per atom in the atmosphere.

For respective  $\text{CO}_2$  and  $\text{N}_2$  volume concentration values of 0.033% and 78.08% [7], we have

$$N(14) = N(\text{C}) \frac{78.08 \times 2 \times 0.9963}{0.033} = N(\text{C}) \times 4.7 \times 10^3.$$

But

$$N(\text{C}) = \frac{6.02 \times 10^{23}}{12.01} M(\text{C})$$

and

$$N(14) = 2.4 \times 10^{26} M(\text{C}) \text{ cm}^{-3}, \quad (4)$$

where  $N(\text{C})$  and  $M(\text{C})$  are the atmospheric carbon atomic concentration and mass per unit volume respectively. Combining the relations (1), (3) and (4) and using the cross-section value of  $1.82 \times 10^{-24} \text{ cm}^2$  given in Table 1, we obtain the dps/g-of-carbon

$$I(14)/M(\text{C}) = 2.4 \times 10^{26} \times 1.82 \times 10^{-24} \bar{\phi} = 0.25 \quad (5)$$

and therefore

$$\bar{\phi} = 5.7 \times 10^{-4} \text{ n cm}^{-2} \text{ s}^{-1}. \quad (6)$$

The equilibrium number of  $^{39/41}\text{Ar}$  atoms per unit volume is given by

$$N = \frac{N(\text{Ar}) \alpha \sigma_0 \bar{\phi}}{\lambda}, \quad (7)$$

where  $N(\text{Ar})$  is the number of argon atoms per unit volume,  $\alpha$  the isotopic abundance of target atoms, and  $\lambda$  the decay constant of the radioactive atom produced. For  $^{42}\text{Ar}$  a similar expression holds, where in place of  $N(\text{Ar})$  we have the equilibrium number of  $^{41}\text{Ar}$  atoms and  $\alpha = 1$ . By inserting numerical values one obtains:

$$N(39)/N(\text{Ar}) = 3.5 \times 10^{-21} \text{ atoms/Ar atom},$$

$$N(42)/N(\text{Ar}) = 1.5 \times 10^{-42} \text{ atoms/Ar atom},$$

and for the specific activities (given the liquid argon density,  $\rho = 1400 \text{ g/l}$ )

$$I(39) = 6.1 \times 10^{-6} \text{ Bq/l-of-LAr},$$

$$I(42) = 2.1 \times 10^{-26} \text{ Bq/l-of-LAr}.$$

This value is 8 orders of magnitude below the present experimental limit. We can therefore conclude that the natural neutron flux is too low to produce a detectable  $^{42}\text{Ar}$  activity or a serious background for low energy neutrino events from  $^{39}\text{Ar}$  activity.

### 3. Neutrons produced in nuclear explosions

We assume a very short irradiation time, much less than the mean life of the isotopes of interest, so that decay during irradiation can be neglected. We also assume a very

high neutron density, but small enough that there is no appreciable decrease of stable argon from neutron capture, except for  $^{39}\text{Ar}$  whose capture cross section is particularly high.

The number of radioactive atoms produced in a volume element  $dV$ , in a single explosion, corresponding to an irradiation time  $t$ , is given by

$$dN(39) = N(\text{Ar}) \alpha(38) \frac{\sigma(38)}{\sigma(39) - \sigma(38)} \times (e^{-\sigma(38)\phi t} - e^{-\sigma(39)\phi t}) dV, \quad (8)$$

$$dN(42) = N(\text{Ar}) \alpha(40) \sigma(40) \sigma(41) \frac{(\phi t)^2}{2} dV, \quad (9)$$

where  $\alpha(x)$  and  $\sigma(x)$  are the isotopic abundance and the mean cross-section for neutron capture by  $^x\text{Ar}$ , and  $\phi$  is the neutron flux. The above equations would allow an easy calculation of the nuclear explosions contribution to  $^{39/42}\text{Ar}$  activity if a few parameters were known, e.g. neutron flux levels, spatial and energy distribution, mean cross sections, the number of nuclear explosions, the energy yield, etc. Although such information is not available we nevertheless believe that it is possible to derive a sound estimate of the  $^{39}\text{Ar}$  concentration and of an upper limit for the  $^{42}\text{Ar}$  concentration.

We refer again to the  $^{14}\text{C}$  situation, for which the measured mean increase in concentration caused by nuclear explosions is between 1.5 [5] and 2 [9]. It is reasonable to suppose that the increase of  $^{39}\text{Ar}$  activity is nearly the same, since the production cross-sections have substantially the same behaviour as a function of energy. Hence, the contribution to  $^{39}\text{Ar}$  concentration due to nuclear explosions is at most:

$$N(39)/N(\text{Ar}) = 3.5 \times 10^{-21} \text{ atoms/Ar atom}.$$

We can now derive an estimate of the  $N(42)/N(39)$  ratio. The energy release from one kton of TNT is about  $4 \times 10^{12} \text{ J}$ . In a D–T fusion event one neutron and 17 MeV of energy are released, so that  $1.5 \times 10^{24}$  neutrons are produced in a one kton thermonuclear explosion. These neutrons will propagate as a spherical wave and the fluence ( $\phi t$ ) will decrease approximately as:

$$\phi t = \frac{1.5 \times 10^{24}}{4 \pi r^2} e^{-r/L} \text{ n/(cm}^2 \text{ kton)}, \quad (10)$$

with  $L \approx 2 \times 10^4 \text{ cm}$  [10].

Let us consider a nuclear weapon having a yield equal to  $P$  kton; the total number of  $^{39/42}\text{Ar}$  atoms produced is

$$N(39/42) = \int_{d_1}^{d_2} dN(39/42), \quad (11)$$

where  $d_2$  is the distance up to which the neutron flux is significant and  $d_1$  is some distance near to the explosion point. We suppose  $d_1 \ll L$  and  $d_2 \gg L$ . With these conditions and considering that in most of the space around the

explosion point  $e^{-\sigma(38)\Phi t} = 1$  and  $e^{-\sigma(39)\Phi t} = 1 - \sigma(39)\Phi t$ , we obtain

$$\begin{aligned} N(39) &\approx N(\text{Ar})\alpha(38)\sigma(38) \times 1.5 \times 10^{24}LP \\ &= 15N(\text{Ar})P, \end{aligned} \quad (12)$$

$$\begin{aligned} N(42) &\approx N(\text{Ar})\alpha(40)\sigma(40)\sigma(41)\frac{P^2}{2} \times (1.5 \times 10^{24})^2 \\ &= 0.36N(\text{Ar})P^2. \end{aligned} \quad (13)$$

In most of the experiments, the nuclear weapons yield was below 1 Mton. If we take as an upper limit 1.5 Mtons in 90% of the explosions, and 15 Mtons in the remaining 10% [11], we have from Eqs. (12) and (13)  $\bar{N}(42)/\bar{N}(39) = 0.21$  so that

$$\begin{aligned} N(42)/N(\text{Ar}) &= 0.21 \times 3.5 \times 10^{-21} \\ &= 7.4 \times 10^{-22} \text{ atoms/Ar atom}, \end{aligned} \quad (14)$$

which is 20 orders of magnitude larger than the estimate from naturally occurring neutron captures.

#### 4. Conclusions

We can conclude from the above calculation that if  $^{42}\text{Ar}$  is present in an appreciable amount it has been produced exclusively in nuclear explosions and that the previously stated experimental upper limit [3] is probably at least  $10^3$  times higher than the real concentration.

We will try to obtain a direct experimental confirmation from a new measurement at the Gran Sasso Laboratory, using a bigger germanium detector and an improved experimental set-up with respect to the one described in

Ref. [3a], to establish firmly the corresponding background for low energy neutrino events in the ICARUS experiment.

If the computed  $^{42}\text{Ar}$  concentration will be confirmed, we can conclude that this background is not a serious problem for the detection of low energy neutrino events in the ICARUS experiment.

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