REASSESSMENT OF THE EVIDENCE FOR ^{36}Ar FROM ^{36}Cl DECAY IN ALLENDE SODALITE.

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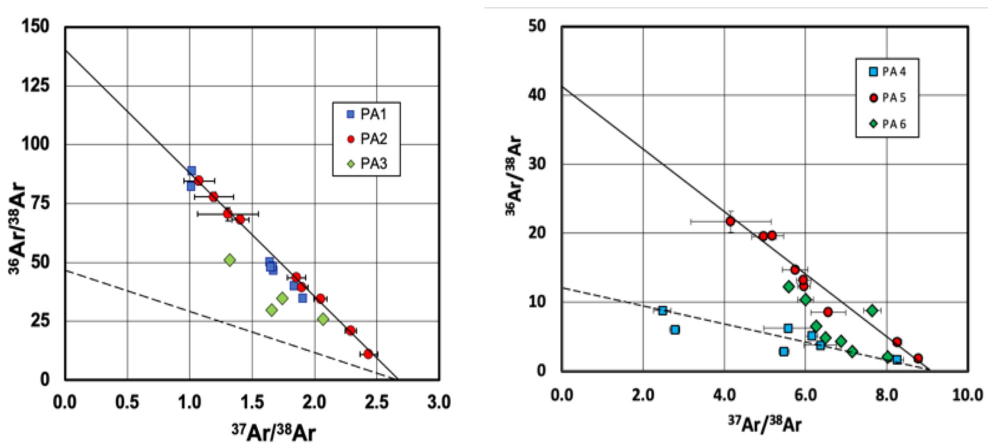
Introduction: A reassessment of the published evidence[1] for excess ^{36}Ar from ^{36}Cl decay in Allende sodalite, based on isotope mixing calculations, supports the former presence of ^{36}Cl with a ^{36}Cl/^{35}Cl ratio of $1.9\text{x}10^{-8}$ to $2.2\text{x}10^{-8}$. While most data points, figures below, lie on correlation lines corresponding to excess ^{36}Ar from ^{36}Cl decay, some define a lower line with an intercept expected for irradiation by cosmogenic neutrons over the last 5.2 Myrs, i.e. from sodalite which lost its excess ^{36}Ar or never had any. The implication that the ^{36}Cl was introduced live into the sodalite ~7Ma after CAI formation places serious constraints on the timing of possible early solar system models combined with neutron fluence requirements. A lower limit on the neutron flux requirement, 23 times the current cosmogenic flux, is possible but only if the sodalite formation coincided closely with the end of neutron irradiation.

Isotope mixing calculations: For a mixture (C) of two isotope ratios (A) and (B) the fraction f[B]of element B in the mixture is given by f[B] = [(A) – (C)]/[(A) – (B)]. Applying this expression to (^{36}Ar/^{38}Ar), the intercept in figs. 1a and 1b, we obtain: $(^{36}\text{Ar}/^{38}\text{Ar})_A$ = (^{36}Ar/^{38}Ar)/(1 - $[^{38}\text{Ar}]_B/[[^{38}\text{Ar}]_B + [^{38}\text{Ar}]_A]$). Subscript A refers to Ar produced by cosmogenic neutron irradiation plus any additional ^{36}Ar component. Subscript B refers to Ar isotopes produced by the reactor neutron irradiation. Note that $(^{36}\text{Ar}/^{38}\text{Ar})_B = 0$, reflecting the long half-life of ^{36}Cl produced. The two unknowns are $(^{36}\text{Ar}/^{38}\text{Ar})_A$ and $[^{38}\text{Ar}]_A$. $[^{38}\text{Ar}]_B$ is the amount of ^{38}Ar produced from ^{37}Cl in each of the reactor irradiations, $(5.43 \pm 0.54)\text{x}10^{-10}$ and $(2.00 \pm 0.05)\text{x}10^{-9}$ respectively. (^{36}Ar/^{38}Ar) is the value of the intercept for each of figs 1a and 1b, 139 ± 2 and 42.5 ± 1.0. Substituting the known values and solving the two simultaneous equations we obtain, $[^{38}\text{Ar}]_A/[^{37}\text{Cl}] = (1.06 \pm 0.11)\ \text{x}10^{-10}$ and $(^{36}\text{Ar}/^{38}\text{Ar})_A = 855 \pm 78$. The proportion of $(^{36}\text{Ar}/^{38}\text{Ar})_A$ which can be assigned to cosmogenic neutron production is $\sigma_{35}/\ \sigma_{37}\ .\ ^{35}\text{Cl}/^{37}\text{Cl}.\ (1 - \tau_{36}/T_{exp})\ .\ B_{36}$, where σ_{35} and σ_{37} are the neutron capture cross sections, τ_{36} the mean life of ^{36}Cl, T_{exp} the cosmic ray exposure duration of Allende, 5.2 Ma, and B_{36} the fraction of ^{36}Cl which decays to ^{36}Ar, 98%. Based on estimates of $\sigma_{35}/\ \sigma_{37}$ from 68 to 100, (^{36}Ar/^{38}Ar) from cosmogenic neutron irradiation is in the range 200 to 290, and $[^{36}\text{Ar}]/[^{35}\text{Cl}] = (0.69\text{ to }1.00)\text{x}10^{-8}$. The remaining excess ^{36}Ar corresponds to (^{36}Ar/^{38}Ar) from 565 to 655. Relative to ^{35}Cl this indicates $[^{36}\text{Ar}]/[^{35}\text{Cl}] = 1.9\text{x}10^{-8}$ to $2.2\text{x}10^{-8}$, which is consistent with the value published previously for (^{36}Cl/^{35}Cl)[1].

Lower intercept: A question, not considered by Turner et al[1], concerns the significance of data points below the main correlation lines in figures 1a and 1b. In particular the data from sodalite PA4 defines a separate correlation and intercept, indicated by the dashed line in fig.1b. Especially noteworthy is the intercept, (^{36}Ar/^{38}Ar) ~13, which is as expected from recent cosmic ray irradiation alone. This indicates that some areas of sodalite have lost or never contained excess ^{36}Ar from ^{36}Cl and may be a feature of previous unsuccessful attempts to locate ^{36}Ar from ^{36}Cl decay.

Modelling, timing and neutron fluxes: The absence of neutron generated ^{128}I and ^{38}Ar, above the cosmogenic level, was the sine qua non used to identify ^{36}Cl as the source of the excess ^{36}Ar. A direct implication is that live ^{36}Cl, with (^{36}Cl/^{35}Cl) ~ $2\text{x}10^{-8}$ was incorporated into the sodalite sometime after the end of the neutron irradiation. I-Xe dating indicates that the sodalite formed 7 million years after the host CAI[1]. The level of (^{36}Cl/^{35}Cl) produced by a uniform neutron irradiation of a Cl bearing fluid is: $N = P/\lambda.(1 - \exp(-\lambda T_1).\exp(-\lambda T_2)$, T_1 the duration of the irradiation, T_2 the time after. $P = (\sigma_{35}.\Phi)$. Φ is the neutron flux and σ_{35} the ^{35}Cl capture cross section. Solutions with P > 23 times the cosmogenic flux are required but neither the actual value nor the irradiation environment are currently known.



References: [1] Turner G., et al., (2013) *Geochimica et Cosmochimica Acta* 123:358-367.

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