

SEARCH FOR EXTINCT CHLORINE-36 IN VIGARANO CAIs. D. Nakashima¹, U. Ott¹, P. Hoppe¹, and A. El Goresy². ¹Max-Planck-Institut für Chemie, J.-J.-Becher-Weg 27, 55128 Mainz, Germany. (naka@mpch-mainz.mpg.de), ²Bayerisches Geoinstitut, Universität Bayreuth, 95447 Bayreuth, Germany.

Introduction: Now extinct short-lived radionuclides with half lives of 10^5 to 10^8 years may serve as sensitive chronometers and tracers for processes in the early solar system and can provide constraints for models of nucleosynthesis. Radioactive ^{36}Cl ($T_{1/2} = 3 \times 10^5$ years) has been suspected to have been present in the early solar system. Its actual detection, however, has proven to be difficult [e.g., 1]. Part of the problem is that Cl-rich phases such as sodalite tend to be alteration rather than primary phases and that the decay of ^{36}Cl primarily leads to volatile ^{36}Ar (98.1%; β^- decay). There has been no clear evidence about ^{36}Ar excesses induced by ^{36}Cl decay in sodalite of Ca-Al-rich inclusions (CAIs), which may be due to ^{36}Ar loss. Although only 1.9% of decays lead to ^{36}S (β^+ decay and electron capture), it was found that ^{36}S excesses correlate with Cl/S in sodalite from a CAI in the Ningqiang meteorite, corresponding to $^{36}\text{Cl}/^{35}\text{Cl} \sim 5 \times 10^{-6}$ at the time of sodalite formation [2]. A similar ratio was found in sodalite of the Pink Angel CAI from the Allende meteorite [3, 4]. On the other hand, we found no evidence for extinct ^{36}Cl in our study of another fine-grained Allende CAI [5]. Here we report the results of Cl- and S-isotopic analysis of sodalite in CAIs from the Vigarano meteorite.

Experimental: Two thick sections were prepared from Vigarano and searched for the occurrence of sodalite in CAIs using a FE-SEM at Bayreuth University. Back-scattered electron (BSE) imaging was used for SEM studies. Elemental compositions of constituent minerals of the CAIs were subsequently measured using an EPMA equipped with WDS at Mainz University. WDS quantitative analyses were performed at 15kV accelerating voltage and 10nA beam current with a focused beam $1\mu\text{m}$ in diameter. Isotopic analysis of Cl and S in sodalite was by the NanoSIMS at Max-Planck-Institute for Chemistry in Mainz. Sulfur isotopes 32, 34, and 36 as well as ^{37}Cl were measured in multicollection ion counting as negative ions produced by a Cs^+ primary beam of $\sim 25\text{pA}$ and $\sim 1\mu\text{m}$ in diameter. The ^{36}S count rate was corrected for background measured at 14 millimasses below the ^{36}S peak. Obtained $^{37}\text{Cl}/^{34}\text{S}$ ratios were converted into $^{35}\text{Cl}/^{34}\text{S}$ ratios using a ratio of 3.13 for $^{35}\text{Cl}/^{37}\text{Cl}$ [6]. In addition, $^{35}\text{Cl}/^{34}\text{S}$ ratios were corrected with a relative sensitivity

factor of 0.83 (favoring Cl) as determined by [3]. Mundrabilla troilite and FeS grains close to the CAIs were used as standards.

Results and discussion: In the Vigarano thick sections, we found two sodalite-bearing CAIs (VI-1 and VI-2), $\sim 2\text{mm}$ and $\sim 1.2\text{mm}$ in size. The CAIs are irregularly shaped and rimmed by monomineralic layers of, from innermost to outermost, spinel ($\sim 10\mu\text{m}$ wide), fassaite ($\sim 2\mu\text{m}$ wide), and diopside ($\sim 8\mu\text{m}$ wide). Small CAI fragments occur close to the two main CAIs. The interiors of the main CAIs and the fragments consist mainly of coarse-grained gehlenitic melilite ($20\text{-}500\mu\text{m}$; $\text{Åk } 2\text{-}21$) and small spinel grains ($5\text{-}20\mu\text{m}$). It is considered that the CAIs are fluffy Type A's [7].

Sodalite and nepheline grains ($5\text{-}20\mu\text{m}$) occur as mixtures along cracks of melilite (Fig. 1), suggesting replacement of melilite by sodalite and nepheline via reaction with nebular gases bearing Na, Cl, and K. Although most of spinel grains in the CAIs are rich in Mg and Fe-free (Mg#95-99), spinel grains near the altered regions are rich in Fe (Mg#63-90; Fig. 1). It is considered that Fe was also incorporated into the CAIs during the alteration.

Sodalite and nepheline occur in the main CAIs, but are absent from the small CAI fragments. If alteration occurred after fragmentation, also the CAI fragments should contain sodalite and nepheline. Hence it is likely that the alteration occurred before fragmentation. Since fragmentation should have occurred on the parent body, sodalite and nepheline in the main CAIs may have formed before accretion to the parent body (i.e. in the solar nebula). Similarly, Hsu et al. [3] concluded from the lack of sodalite in Allende matrix surrounding the Pink Angel CAI that alteration in the nebula was more likely. In case of Allende, however, we found numerous sodalite grains in matrix [4, 5], which would argue for parent body alteration instead. Thus, the possibility of multiple alteration episodes should be considered.

Figure 2 shows a $^{36}\text{S}/^{34}\text{S}$ - $^{35}\text{Cl}/^{34}\text{S}$ plot for sodalite in the Vigarano CAIs. Most of the data show no ^{36}S excess and are distributed along the baseline ($^{36}\text{S}/^{34}\text{S} \sim 0.00346$). Only two data points are indicative of ^{36}S excesses, however, with large errors. If taking 2σ errors, the excesses have no statistical significance. Thus, sodalite of the Vigarano CAIs shows no clear ^{36}S

excess. It is possible that the ^{36}Cl - ^{36}S system in sodalite was disturbed by later parent body processes. However, it is inferred from abundances of presolar grains and from Raman spectroscopy of organic material that Vigarano is less altered than Allende [8], which contains CAI-sodalite showing clear ^{36}S excesses [3, 4]. Therefore, it is unlikely that the absence of ^{36}S excess from the Vigarano CAIs is due to later disturbance by parent body processes.

A weighted fit gives no positive correlation between $^{36}\text{S}/^{34}\text{S}$ and $^{35}\text{Cl}/^{34}\text{S}$. An upper limit of $^{36}\text{Cl}/^{35}\text{Cl}$ ratio is estimated to be 1.7×10^{-6} from the 2σ uncertainty of the slope of the regression line, which is clearly lower than the previous results [2, 3].

Lin et al. [2] and Hsu et al. [3] estimated initial $^{36}\text{Cl}/^{35}\text{Cl}$ (i.e. at CAI formation) at $>10^{-4}$. This can be explained by spallation reactions only [3], similar to the case of ^{10}Be [9]. However, ^{10}Be appears to be present in roughly equal abundance in all studied CAIs [e.g., 10], whereas our results (and also [11]) show that ^{36}Cl appears to be variable. There seems to be no simple relationship between ^{10}Be and ^{36}Cl . A larger database of ^{36}Cl is required to establish the relation to other radionuclides.

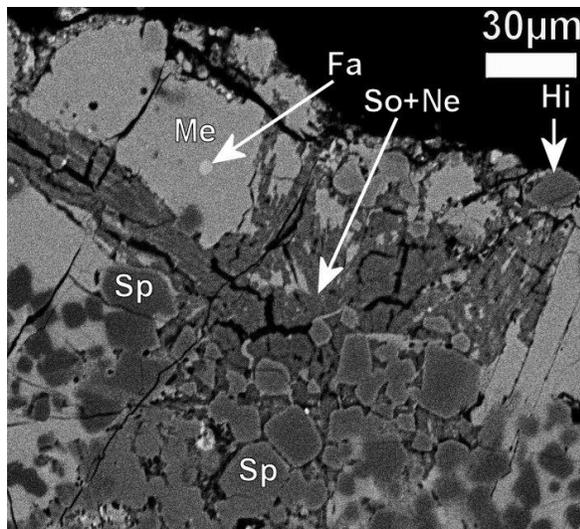


Fig. 1: BSE image of VI-1, showing replacement of melilite (Me) by sodalite (So) and nepheline (Ne). Spinel grains (Sp) near the altered region are also affected by alteration such as incorporation of Fe. The CAI includes also small fassaite grains (Fa) and hibonite grains (Hi).

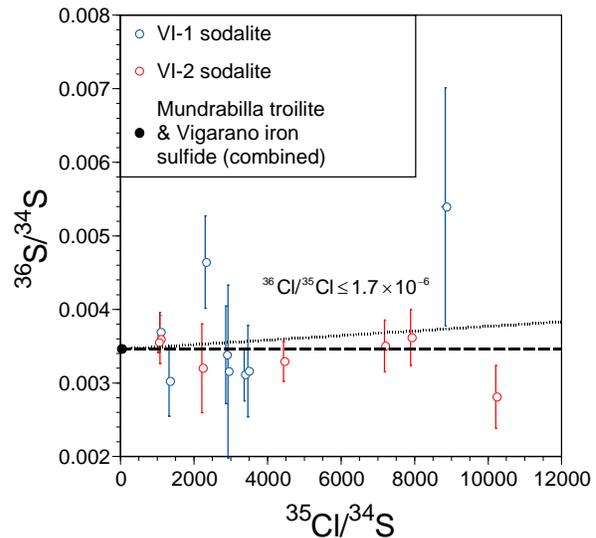


Fig. 2: $^{36}\text{S}/^{34}\text{S}$ vs. $^{35}\text{Cl}/^{34}\text{S}$ plot for sodalite in the Vigarano CAIs, VI-1 and VI-2. Error bars are 1σ and based on Poisson statistics only. The horizontal line corresponds to normal $^{36}\text{S}/^{34}\text{S}$ as deduced from standards ($^{35}\text{Cl}/^{34}\text{S} \sim 0$). A weighted fit line yields a 2σ upper limit of 1.7×10^{-6} for the $^{36}\text{Cl}/^{35}\text{Cl}$.

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