SEARCH FOR EXTINCT CHLORINE-36 IN AN ALLENDE CAI. M. Plagge, U. Ott and P. Hoppe, Max-Planck-Institut für Chemie, Becherweg 27, D-55128 Mainz (ott@mpch-mainz.mpg.de).

Introduction: The search for the former existence in the Early Solar System of the now extinct radionuclide ^{36}Cl ($T_{½}$ = 3x10^{5} a) has proven to be difficult [e.g., 1]. This is at least partly due to the volatility of chlorine and the fact that in primitive meteorites Cl occurs in high abundance only in alteration phases such as sodalite ($Na_8Al_6Si_6O_{24}Cl_2$). 36-Cl decays to both ^{36}Ar by β^- decay (98.1 %) and to ^{36}S by β^+ decay and electron capture (1.9 %). Early attempts to establish its presence focused on ^{36}Ar in fine-grained Ca-Al-rich inclusions of the Allende CV3 meteorite that contain several thousand ppm of Cl in bulk [2]. Excesses of ^{36}Ar observed at low to intermediate gas extraction temperatures, with ^{36}Ar/^{38}Ar ranging as high as ~90 [2,3], if interpreted as due to extinct ^{36}Cl, correspond to ^{36}Cl*/^{35}Cl ratios on the order of ~ 10^{-8}, but could be explained as due to neutron irradiation during the ~5 Ma recent cosmic ray exposure of the meteorite.

A positive result was reported by [4] who observed apparent ^{36}Ar overabundances in bulk measurements of the CV3 meteorite Efremovka. Their inferred ^{36}Cl*/^{35}Cl ratio of ~1.4x10^{-6} has been widely cited in the subsequent literature and used in models that try to provide a coherent model for the origin of extinct radionulcides (e.g., [5, 6]), but has been shown to have arisen from an analytical artifact [7].

Only recently there appears to be unambiguous evidence for the presence of ^{36}Cl in the ESS. This is based on excesses of ^{36}S observed by SIMS that correlate with Cl/S ratio in a CAI from the Ningqiang anomalous carbonaceous chondrite [8] and, more recently in the “Pink Angel” inclusion as well as a porphyritic olivine chondrule in Allende [9]. From a ^{36}Cl*/^{35}Cl ratio of ~4x10^{-6} at the time of sodalite formation the authors infer a ratio at the time of CAI formation of >10^{-4}. Using the NanoSIMS with its higher sensitivity and multi-collection capability we have begun a study of the Cl-S system in CAIs hoping to confirm the results of [8, 9] with higher accuracy.

Sample: A thick section was prepared from the Allende meteorite containing a “typical” fine-grained inclusion (CAI MP-1) similar to those studied by [2] for Cl abundance and noble gases. Seven areas of some 200 to 700 µm size within the CAI were mapped for Cl abundances leading to the identification of numerous grains of sodalite, with a typical size ~20 µm (Figs. 1, 2). Notably, we also found sodalite grains in Allende matrix, contrary to the case of [9] who did not observe sodalite in Allende matrix nor in a second chondrule close to the Pink Angel.

Fig. 1. Optical microscope picture showing regions in CAI MP-1 searched for sodalite. FoV ~ 4 mm x 3 mm.

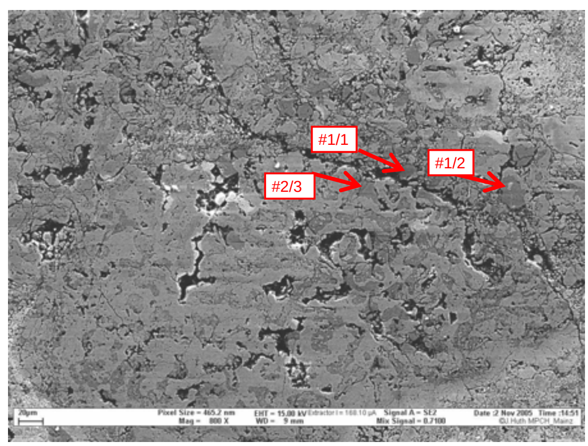

Fig. 2. SEM picture showing areas #1 and #2 in more detail. Analyzed sodalite grains are marked by arrows.

NanoSIMS Analyses: SIMS analyses reported here were performed on grains #1 and #2 in area #1 and on grain #3 in area #2. 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 ~25 pA. After the first two measurements on grain #2 in area #1 seemed to indicate excesses of ^{36}S, we noted the presence during sodalite analyses of a broad, small, but nevertheless non-negligible background in the ^{36}S mass region, probably due to scattered ions and usually decreasing during the course (several hours) of our measurements. The remaining 4 analyses reported here were performed in a “combined mode". After each measuring cycle of 5 seconds duration background data for correction were taken for another 5 seconds at a peak position 14 millimasses below that of ^{36}S using the same detector as for the ^{36}S measurements proper. Interspersed with the sodalite measurements were analyses of low-Cl regions in the CAI as well as FeS from the Mundrabilla iron meteorite. The latter served for calibrating the relative efficiencies of our detectors, assuming Mundrabilla troilite to have the same S isotopic composition as the Canyon Diablo troilite standard [$^{34}S/^{32}S$ = 0.044149; $^{36}S/^{32}S$ = 0.0001534; [10]).

Results: Under our measurements conditions count rates at sodalite were ~ 10^5 cps for ^{37}Cl. Relevant results are summarized in Fig. 3, where errors shown are based on Poisson statistics only. Also shown are the data of [8] and [9]. To allow comparison we use a diagram of $^{36}S/^{34}S$ vs. $^{35}Cl/^{34}S$. Mass fractionation correction of $^{36}S/^{34}S$ relative to the standard was performed based on measured $^{34}S/^{32}S$. A normal ratio of 3.13 for the $^{35}Cl/^{37}Cl$ [11] was used for converting ^{37}Cl into ^{35}Cl abundances. In addition a relative sensitivy factor of 0.83 (favoring Cl) was assumed, as determined by [9]. Following [9], the Ningqiang Cl/S ratios of [8] were recalculated using the same sensitivity factor.

Resulting $^{35}Cl/^{34}S$ ratios in our sodalite measurements range up to ~29,000. While one of the lower Cl/S data points (sodalite 1/1 at $^{35}Cl/^{34}S$ ~9000) is compatible with an enhanced $^{36}S/^{34}S$ as in [8, 9], this is not supported by the results for higher Cl/S. We estimate a conservative upper limit for the ratio $^{36}S_{excess}/^{35}Cl$ of $1x10^{-8}$, which translates into an upper limit for $^{36}Cl^*/^{35}Cl$ of $5x10^{-7}$, an order of magnitude lower than the ratio inferred in [8, 9].

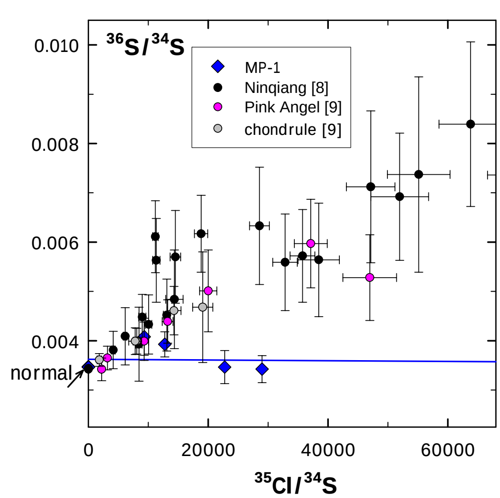

Fig. 3: Plot of $^{36}S/^{34}S$ vs. elemental ratio $^{35}Cl/^{34}S$. In our data (blue) there is no evidence for enhanced ^{36}S.

Discussion: Sodalite is a volatile-rich alteration product, not a primitive high temperature condensate and thus does not date the formation of the CAI proper (cf. [9]). An open question is whether the alteration occurred in the solar nebula or on the meteorite parent body. Hsu et al. [9] conclude from the lack of sodalite in a second chondrule close to the Pink Angel and in the matrix of Allende that alteration in the nebula is more likely; however, in our search in Allende we also found numerous sodalite grains in the matrix. Consideration of textures indicates that sodalites in the Ninqiang CAI may be of a more primitive nature, i.e. may be more likely to have formed during alteration in the nebula than those in our CAI, but this conclusion is not firm [8]. Textures of sodalites in CAI MP-1 are essentially identical to those in the Pink Angel.

While originally a supernova source has been suggested for the $^{36}Cl^*$ observed in Ninqiang [8], according to arguments presented in [1, 9] an intense late irradiation process in the Early Solar System may be a more likely source that possibly also produced the observed extinct $^{10}Be^*$. In order to further evaluate this, it will be necessary to establish a relationship between the occurrences of both extinct radionuclides. We note, however, that, unlike $^{26}Al^*$ e.g., $^{10}Be^*$ appears to have been present in roughly equal abundance in all CAIs that have been studied [12, 13]. The lack of observable $^{36}Cl^*$ in CAI MP-1 then suggests that there is not a 1:1 relationship between the two nuclides. What is clearly needed now, is a larger database for $^{36}Cl^*$. Possibly there have been several episodes of alteration distinct in time and maybe location as well.

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