EVIDENCE FOR LIVE ^{36}CL IN CA-AL-RICH INCLUSIONS FROM THE NINGQIANG CARBONACEOUS CHONDRITE.

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Introduction: The short-lived radionuclide ^{36}Cl decays to either ^{36}Ar (98.1%, β^-) or ^{36}S (1.9%, ε and β^+), with a half life of 3.01×10^5 yr [1]. Both the nucleosynthetic [2] and spallation [3] models suggest high initial ^{36}Cl/^{35}Cl ratios ((^{36}Cl/^{35}Cl)$_o$ up to ~10^{-4}) in the early solar system. Previous observed excess ^{36}Ar in Efremovka matrix has been interpreted to represent a much lower (^{36}Cl/^{35}Cl)$_o$ ratio of ~1×10^{-6} [4]. From the observed ^{36}S excesses in sodalite in calcium-aluminum-rich inclusions (CAIs), we report in this study the first direct evidence of the presence of ^{36}Cl in primitive meteorites. The inferred (^{36}Cl/^{35}Cl)$_o$ ratios range from ~5×10^{-6} to ~1×10^{-5}.

Samples and Experimental: Sodalite ($Na_8Al_6Si_6O_{24}Cl_2$) in CAIs from Ningqiang (CV3), EET 87746 (EH3) and ALH 77295 (EH3), and coarse-grained djerfisherite ($(K,Na)_6(Fe,Ni,Cu)_{25}S_{26}Cl$) in Qingzhen (EH3) were analyzed for this study. One of the CAIs (J1-1#1) measured in this work is anorthite-spinel-rich inclusion (ASI) in the Ningqiang carbonaceous chondrite [5], which consists of a spinel-rich core, a spinel-anorthite-Ca-pyroxene mantle, and a melilite-spinel crust. This inclusion was formed by alteration of a Type A inclusion, with melilite replaced by anorthite and Ca-pyroxene. The bulk composition of the ASI is identical to those of Type C inclusions, hence probably precursors of the latter [5]. Sodalite and nepheline, both replacing anorthite, coexist in the mantle of the ASI. The two CAIs in ALH 77295 and EET 87746 consist mainly of spinel, pyroxene, sodalite, and nepheline.

Sulfur isotopes (^{33}S, ^{34}S, and ^{36}S) and ^{35}Cl of the samples were measured with the ASU Cameca ims-6f ion microprobe. An O^- primary beam of ~0.1nA at 12.5 keV was focused to <10 μm in size. Secondary ions were accelerated to +9 keV and collected with an electron multiplier. Transfer lenses were tuned for a 75 μm imaged field to increase the secondary ion transmission. Hydride and other molecular interferences to the S isotopes were eliminated under high mass resolution conditions with a mass resolving power ($m/\Delta m$) of 4300. Troilite and djerfisherite in Qingzhen were analyzed as standards for S isotopes. ^{36}S/^{34}S ratios are corrected for instrumental mass fractionation internally using ^{33}S/^{34}S. The relative sensitivity factor of ^{35}Cl/^{34}S was determined using djerfisherite from Qingzhen.

Results and Discussion: Four assemblages of sodalite and nepheline with high ^{35}Cl/^{34}S ratios (up to 57,000) in the ASI have been analyzed. All of them show clear excesses of ^{36}S that are correlated with ^{35}Cl/^{34}S ratios (e.g., Fig. 1). The inferred (^{36}Cl/^{35}Cl)$_o$ ratios are $(5.0 \pm 0.6)\times10^{-6}$, $(5.1 \pm 1.0)\times10^{-6}$, $(7.7 \pm 2.5)\times10^{-6}$, and $(1.1 \pm 0.2)\times10^{-5}$. Because of low ^{35}Cl/^{34}S ratios (<100) resulting from contamination from fine-grained sulfide (probably troilite), no resolvable ^{36}S excess was detected in the two sodalite-bearing CAIs in EET87746 and ALH 77295. The low ^{35}Cl/^{34}S ratios of djerfisherite in Qingzhen also prevent possible resolvable observation of ^{36}S excess.

The inferred (^{36}Cl/^{35}Cl)$_o$ ratios from our study are about 4 to 8 times higher than that ($(1.4 \pm 0.2)\times10^{-6}$) estimate based excess ^{36}Ar [4]. Two reasons may account for the difference. First, in addition to contribution from the decay of ^{36}Cl, most ^{36}Ar in meteorites is a mixture of a trapped component, a spallation component, and decay of cosmogenic ^{36}Cl. Estimation of excess ^{36}Ar from the now-extinct nuclide ^{36}Cl is highly based on subtraction of these components, especially the most abundant trapped component. Therefore, large uncertainty could be introduced during the process. Second, the excess ^{36}Ar was observed in the fine-grained matrix material of Efremovka [4], which probably formed later than sodalite in the CAI in the Ningqiang meteorite, hence, it has a lower (^{36}Cl/^{35}Cl)$_o$ value. Systematic study of a large number of CAIs in Ningqiang supports the idea that the alteration reactions that resulted in sodalite formation in its CAIs took place in the solar nebula instead of on its asteroidal parent body [5,6].

The (^{36}Cl/^{35}Cl)$_o$ ratios observed in sodalite obviously cannot represent the "canonical" value when CAIs first formed in the solar nebula. To better constrain the (^{36}Cl/^{35}Cl)$_o$ values in the early solar nebula, we need to combine ^{36}Cl-^{36}S analysis with other isotopic systems, such as ^{26}Al-^{26}Mg, which will be analyzed in the near future. Nonetheless, petrological observation can also help us evaluate the (^{36}Cl/^{35}Cl)$_o$ evolution in the solar nebula. Textural relationships in the ASI suggests a formation sequence of condensation of fluffy Type A CAIs, followed by high temperature alteration of melilite to produce anorthite and Ca-pyroxene, a separate heating event, and then low temperature alteration of anorthite to produce sodalite and nepheline [5]. The low temperature alteration took place after formation of Type C inclusions that have initial ^{26}Al/^{27}Al ratios of ~1×10^{-5} [7]. Assuming a time difference of 1.5 Ma (2 times of the half-life of ^{26}Al) between the low temperature alteration and formation of fluffy Type A CAIs (with an initial ^{26}Al/^{27}Al ratio of 5×10^{-5}), we can infer the (^{36}Cl/^{35}Cl)$_o$ ratio at the beginning of CAI formation to be (~2-4)$\times10^{-4}$.

Similar to many other short-lived radionuclides (such as ^{26}Al, ^{53}Mn, and ^{41}Ca), two schools of thought [e.g., 2, 3] about the origin of ^{36}Cl exist: nucleosynthetic introduction or local irradiation. The local irradiation model [3] predicts a (^{36}Cl/^{35}Cl)$_o$ ratio of ~1.3×10^{-4}. Whereas ratios of 3×10^{-6} – 2×10^{-4} were estimated for (^{36}Cl/^{35}Cl)$_o$ in the early solar nebula based on injection of fresh debris from a Type II supernova [2]. Because of the overlap of these predictions, uniquely assessing origin of this nuclide may be difficult. In any case, it will certainly require more analyses and correlation with other short-lived isotopes.

References: [1] Endt P. M. (1990) *Nuclear Physics* A521,1. [2] Wasserburg J. G. et al. (1998) *Ap.J.* 500, L189. [3] Leya I. et al. (2003) *Ap.J.* 594, 605. [4] Murty S. V. S. et al. (1997) *Ap.J.* 475, L65. [5] Lin Y. et al. (1998) *Meteoritics & Planet. Sci.* 33, 435. [6] Lin Y. et al. (2003) *GCA* 67, 2251. [7] MacPherson J. G. et al. (1995) *Meteoritics*, 30, 365.

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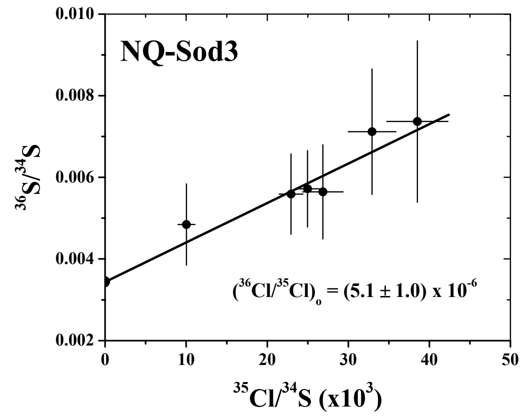

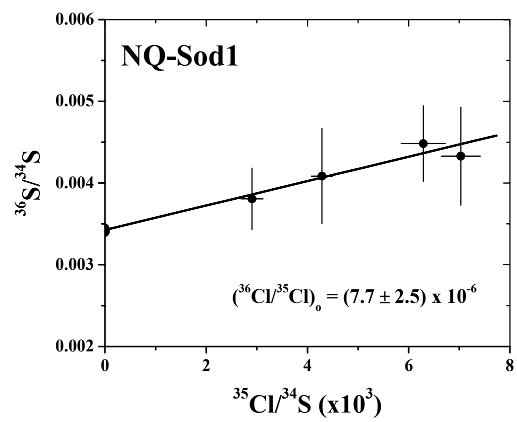

Fig. 1. ^{36}S/^{34}S versus ^{35}Cl/^{34}S plots of sodalite in an anorthite-spinel-rich inclusion (ASI) from the Ningqiang carbonaceous chondrite. Error bars are 2σ.