

³⁶Cl, ²⁶Al AND OXYGEN ISOTOPES IN AN ALLENDE CAI: IMPLICATIONS FOR SECONDARY ALTERATION IN THE EARLY SOLAR SYSTEM. T. Ushikubo¹, Y. Guan¹, H. Hiyagon², N. Sugiura², and L. A. Leshin³, ¹Arizona State University, Department of Geological Sciences, PO Box 871404 Tempe, AZ, 85287-1404, U.S.A. (Takayuki.Ushikubo@asu.edu), ²University of Tokyo, Department of Earth and Planetary Science, 7-3-1 Hongo, Bunkyo, Tokyo, 113-0033, Japan, ³NASA-GSFC, MD, 20771, U.S.A.

Introduction: Ca-Al-rich inclusions (CAIs) from Allende usually have secondary alteration phases such as anorthite, grossular, sodalite, and nepheline [1-3]. Although these phases are considered to have been formed by relatively low temperature reactions between CAIs and a surrounding nebular gas [1, 2], it is not well known that when such processes occurred. ³⁶Cl-³⁶S and ²⁶Al-²⁶Mg systematics are potentially useful for chronologic study on secondary alteration processes. ³⁶Cl decays into ³⁶Ar (98.1%, β⁻) and ³⁶S (1.9%, ε and β⁺) with a half life of 0.3 Myr. ²⁶Al decays into ²⁶Mg with a half life of 0.72 Myr.

To better understand the timing and location of secondary alteration processes in the early solar system, in this study we measured ³⁶Cl-³⁶S and ²⁶Al-²⁶Mg systematics, and O isotopes of an Allende type B2 CAI, CAI#2, using an ion microprobe. Result of Mg isotopic measurements were previously reported in [4].

The Allende CAI studied: CAI#2 is a coarse-grained type B2 inclusion of ~1mm in size from the Allende CV3 chondrite (Figure 1). This CAI consists of fassaite, melilite, spinel, anorthite, grossular, sodalite, and minor Fe-rich phases. There are two distinct alteration regions in CAI#2. One is the Anorthite-Grossular (An-Gr) region that is observed at the grain boundary of melilite or fassaite in the interior of CAI#2 and mostly consists of anorthite(-A) and grossular. The other is the Na-rich region that is observed along the edge of CAI#2 and mostly consists of sodalite, anorthite(-B), and Fe-rich phases.

Analytical conditions: Isotopic measurements were performed using the Cameca ims-6f ion microprobes at Arizona State University and University of Tokyo. Analytical conditions for S isotopes are similar to [5]. We applied a sensitivity factor of 0.69 (with Cl ionizing better), determined using a terrestrial hauyne standard, which contains 5.4 wt.% S and 0.23 wt.% Cl. Analytical conditions for Mg isotopes are similar to [6]. Oxygen isotopes were measured using a +10.0 kV Cs⁺ ion beam of ~0.1 nA, focused into a spot of 10-15μm in diameter in aperture illumination mode. Secondary ions of -9.0 kV were collected with a mass resolving power of ~5000.

Results: Figure 2 shows the ³⁵Cl/³⁴S-³⁶S/³⁴S plot of CAI#2. Significant ³⁶S-excesses were observed in sodalite of CAI#2 and all the data, except for two points, lie on a single line with an inferred

(³⁶Cl/³⁵Cl)₀ = (1.4±0.3)×10⁻⁶. The two highest ³⁵Cl/³⁴S points that show lower ³⁶S-excesses are results of sodalite grains that occur in and around a hedenbergite rich hole. Because the texture (euhedral shape and relatively large, ~40μm in size) of sodalite in the hole is different from other sodalite grains, we consider that its S isotopes could be disturbed by later alteration. Figure 3 shows the ²⁷Al/²⁴Mg-δ²⁶Mg* (²⁶Mg-excesses) plot of CAI#2. Primary phases (spinel, fassaite, and melilite) have low ²⁷Al/²⁴Mg ratios (< 8) and do not yield δ²⁶Mg* larger than 7 ‰. Secondary anorthite(-A) in the An-Gr region, however, shows large δ²⁶Mg* (up to ~35‰), with an inferred (²⁶Al/²⁷Al)₀ of (1.2±0.2)×10⁻⁵. In contrast, secondary anorthite(-B) and sodalite in the Na-rich region contain no significant ²⁶Mg-excess ((²⁶Al/²⁷Al)₀ < 2.0×10⁻⁷). Figure 4 shows O isotopic compositions of CAI#2. All the data points lie on the Carbonaceous Chondrite Anhydrous Minerals (CCAM) line. Oxygen isotopic compositions of spinel and fassaite are highly anomalous (δ¹⁷O ~ δ¹⁸O ~ -55 ‰). Those of melilite and secondary phases in the Na-rich region are less ¹⁶O-enriched. Even though we could not obtain O isotopic composition of individual phases in the An-Gr region, the results of two points on secondary minerals with small amount of primary phases suggest that O isotopic compositions of anorthite and grossular in the An-Gr region are more ¹⁶O-enriched than those of the Na-rich region.

Discussions: *Further evidence of the existence of ³⁶Cl in the early solar system:* Recently, evidence for the existence of ³⁶Cl ((³⁶Cl/³⁵Cl)₀ ~ 4×10⁶) were established in sodalite of CAIs and a chondrule from Ningqiang and Allende [5, 7]. Our results provide further evidence for that. However, the inferred (³⁶Cl/³⁵Cl)₀ of CAI#2 (~1.4×10⁻⁶) is apparently lower than previous results. This could indicate a time difference of sodalite formation or the occurrence of isotopic disturbance after sodalite formed.

Timing of alteration processes: It has been suggested that anorthite and grossular are formed by a reaction between melilite and the nebular gas and that sodalite is formed by a further alteration (introduction of Na and Cl) [1]. This is consistent with the petrologic features of alteration regions in CAI#2. Distinct O and Mg isotopic compositions indicate that the An-Gr region and the Na-rich region are formed by dif-

ferent processes at different time. The observed ^{26}Mg -excess in the An-Gr_s region is much higher than those of primary phases. The higher ^{26}Mg -excesses could not be produced by Mg isotopic exchange between primary phases that had ^{26}Mg -excess and secondary phases that formed after ^{26}Al had already decayed. Our results suggest that some alteration processes occurred while ^{26}Al was alive. Similar observations have been made in other studies of Allende CAIs [8, 9]. Based on the ^{26}Al - ^{26}Mg systematics, the An-Gr_s region formed ~ 1.5 Myr after CAIs formation. In contrast, the Na-rich region could have formed much later (~ 5 Myr) than CAIs.

The ^{16}O -rich isotopic composition and high initial $^{26}\text{Al}/^{27}\text{Al}$ ratios of the An-Gr_s region suggest that a decomposition of melilite (the An-Gr_s region formation) occurred in the solar nebula. Although possible time difference of sodalite formation based on results of ^{36}Cl - ^{36}S systematics seems to be consistent with a nebular origin, there is also a possibility that the ^{36}Cl - ^{36}S systematics were disturbed during later processes. Further studies on ^{36}Cl - ^{36}S systematics of sodalite are required.

References: [1] Hashimoto A. and Grossman L. (1987) *GCA*, 51, 1685–1704. [2] Wark D. A. (1981) *LPS XII*, 1145–1147. [3] Hutcheon I. D. and Newton R. C. (1981) *LPS XII*, 491–493. [4] Ushikubo T. et al. (1999) *Antarctic Meteorite XXIV*, 182–184. [5] Lin et al. (2005) *PNAS*, 102, 1306–1311. [6] MacPherson G. J. et al. (2003) *GCA*, 67, 3165–3179. [7] Hsu et al. (2006) *Ap.J.* (in press). [8] Fagan T. J. et al. (2005) *LPS XXXVI*, Abstract #1820. [9] Fagan T. et al. (2006) *LPS XXXVII*, Abstract #1213.

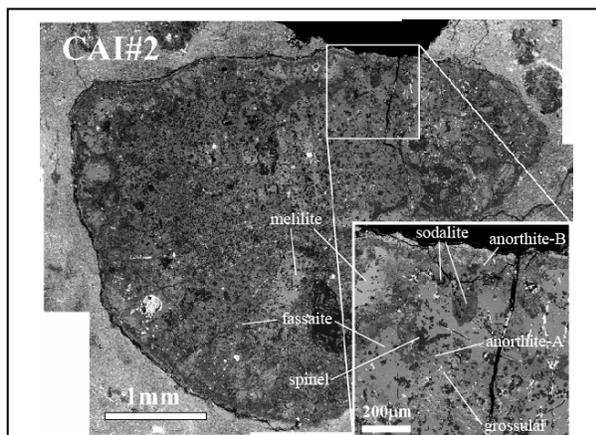


Figure 1. Back-scattered electron image of CAI#2. CAI#2 has two distinct alteration regions, the anorthite-grossular (An-Gr_s) region (along grain boundaries of primary phases) and the Na-rich region (along the edge of this CAI).

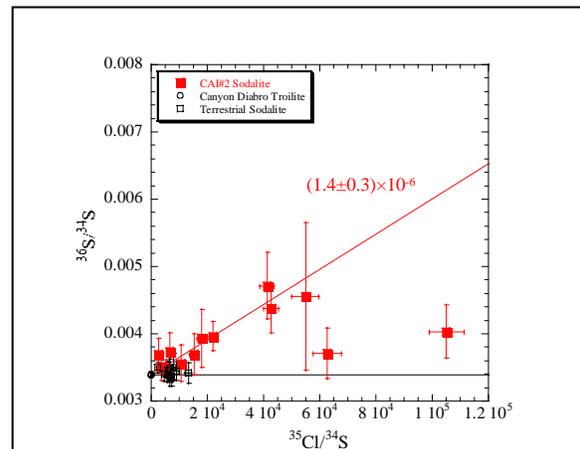


Figure 2. The $^{35}\text{Cl}/^{34}\text{S}$ - $^{36}\text{S}/^{34}\text{S}$ plot of sodalite of the Allende CAI#2.

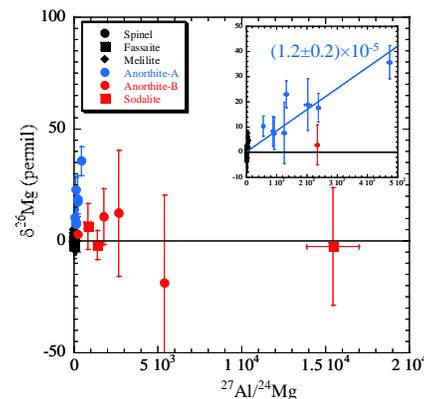


Figure 3. The $^{27}\text{Al}/^{24}\text{Mg}$ - $\delta^{26}\text{Mg}$ plot of CAI#2. Black symbols: primary phases, blue symbols: the An-Gr_s region, and red symbols: the Na-rich region.

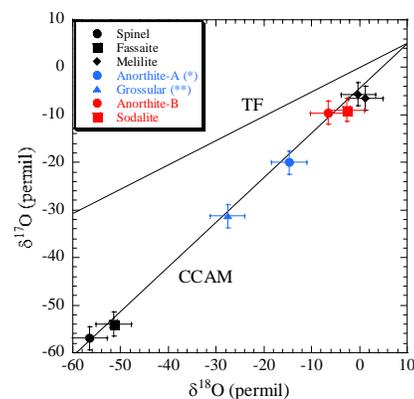


Figure 4. Oxygen isotopic compositions of CAI#2. Black symbols: primary phases, blue symbols: the An-Gr_s region, and red symbols: the Na-rich region. (*) Mixture of $\sim 95\%$ anorthite and $\sim 5\%$ spinel. (***) Mixture of $\sim 80\%$ grossular and $\sim 20\%$ fassaite.