

**SI AND MG ISOTOPE FRACTIONATIONS IN MELILITE IN TYPE B CAIs MEASURED BY SIMS.** N.

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**Introduction:** Si and Mg isotopic compositions are indicators of evaporation processes experienced by CAIs [1]. Bulk Si and Mg isotopic compositions (up to ~5 permil/amu for Si and ~11 permil/amu for Mg) have been reported by [2]. Isotopic heterogeneity within CAIs is of interest because it may reveal how evaporation processes occurred. It may also reveal if secondary processes such as recondensation and equilibration have occurred. Fairly large Mg isotopic fractionations within individual CAIs have been reported [3,4] but it is not known if such Mg isotopic heterogeneity is common among CAIs. Si isotope measurements of CAIs by SIMS are scarce [5]. A recent report [6] suggested that precise and accurate Si isotope measurements may be difficult. Here we report Si and Mg isotope measurements of CAIs by SIMS.

**Samples:** Type B CAIs, two (AL-2 and AL-9) from Allende (CV3) and one (SH-1) from Sahara 98044 (CV3) were examined. AL-2 and SH-1 have well developed melilite mantle whereas AL-9 has a very irregular shape and the melilite mantle is seen only locally. Fassaite is present in the mantle (and/or core) of these CAIs. Anorthite is present in the core (and/or mantle) of AL-9 and SH-1. In AL-2, it is present only near the rim. Melilite compositions in AL-2 and SH-1 are normal, ranging from Ak15 to Ak60 and from Ak20 to Ak43, respectively. Melilite compositions in AL-9 are more gehlenitic (from Ak5 to Ak27) suggesting that this CAI may be akin to type A CAIs. No systematic change in the melilite compositions from the core to the rim was observed in these CAIs.

Crystals of synthetic akermanite and gehlenite and glass samples of Ak50 and Ak30 compositions were used for calibration purposes.

**Ion microprobe measurements:** A Cameca IMS-6F ion probe at Univ. of Tokyo was used for the measurements. Since instrumental isotopic fractionation could result from many factors, exactly the same measurement conditions are required for standards and samples. Therefore, polished sections were prepared in which both a standard sample (glass of Ak50 composition) and a CAI were set. Intrinsic isotope fractionations were obtained from the difference in isotopic compositions between a standard sample and a CAI.

Both Si and Mg isotopic fractionations were measured with the same ion probe setting except for the entrance slit width which was adjusted to give

nearly the same maximum count rates ( $1 \times 10^5$  c/s). A large and strong oxygen primary beam (35 micrometer in diameter; 1.6nA) was used. The secondary ions were extracted from the central area (11 micrometer in diameter) of the sputtered area. The energy slit was wide open and the largest contrast aperture was selected so that all ions (with various energies and directions) were evenly sampled. Measurements were started after ~20 minutes of presputtering when secondary ion signals became steady. The mass resolving power was set to ~4000 which was enough for eliminating all interferences.

Measurements were made at several locations (near the rim, in the mantle and in the core) in the CAIs. In addition to Mg, Al was measured for chronological purposes. The reference values for magnesium isotopic ratios ( $^{25}\text{Mg}/^{24}\text{Mg} = 0.12663$  and  $^{26}\text{Mg}/^{24}\text{Mg} = 0.13932$ ) are taken from [7]. The reference values for Si have not been well established. For now we use the values ( $^{29}\text{Si}/^{28}\text{Si} = 0.050633$  and  $^{30}\text{Si}/^{28}\text{Si} = 0.033474$ ) reported by [8]. Measured Si isotopic compositions do not plot exactly on the mass fractionation line through the origin on the Si three isotope plot, but rather plot slightly below the mass fractionation line. But this is not a problem because we are only concerned with isotope fractionations along the mass fractionation line.

**Results:** The deviations of the isotopic fractionations for the synthetic samples are mostly due to deviations expected by counting statistics. In one case (Ak50 measured with SH-1), however, the deviation of the Mg isotopic fractionation (1sigma = 1.4 permil) was significantly larger than that (1 sigma = 0.6 permil) expected by counting statistics. Thus, there still remained some instrumental factors that were not well controlled in these measurements. Nevertheless, the errors are generally small enough for the purposes of this study. The instrumental isotopic fractionations are identical for the synthetic melilite samples (Ak100, Ak50, Ak30 and Gehlenite set in the same sample holder) within the respective measurement errors for both Si and Mg. Thus isotopic fractionations in CAI melilites (after correcting for a standard melilite) are attributable to intrinsic fractionation (e.g. due to evaporation) rather than matrix effects. The instrumental Mg isotopic fractionation for Ak50 glass set in different holders on different days ranged from -6.9 to -9.0 permil/amu. This is quite similar to those reported by [3]. But the agreement may be fortuitous because the data by [3]

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were obtained from the early stage of sputtering whereas our data were obtained after a long presputtering period. The instrumental Si isotope fractionation for Ak50 glass samples set in different holders on different days ranged from -33.9 to -36.2 permil/amu. This is quite different from that (-49.2 permil/amu) reported by [3]. Such a difference is not unexpected because instrumental mass fractionations depend on the setting of the ion microprobe quite sensitively.

The Si and Mg isotopic compositions within one CAI are nearly the same. The deviations of Si isotope fractionation (1 sigma  $\sim$  1.2 permil/amu) for AL-9 and SH-1 are somewhat larger than that expected from counting statistics. This could be due to intrinsic fractionation within CAIs. But in view of the example shown above that a deviation that is larger than expected by counting statistics could happen even for standard samples, we consider that these could be due to instrumental fractionation. The Si isotopic compositions are not correlated with Mg isotopic compositions, supporting the view that the deviations in the Si isotopic compositions in AL-9 and SH-1 are not intrinsic fractionation. Also, the isotopic compositions do not depend on the location within the CAIs. Deviations of Mg isotopic compositions in three CAIs and Si isotopic compositions in AL-2 are small (1 sigma  $<$  1 permil), not much larger than those expected from counting errors. Thus we consider that both Si and Mg isotopic compositions in the three CAIs are essentially constant. The average Si and Mg isotopic fractionations (and 1 sigma errors) are  $F(\text{Si}) = 2.6 \pm 0.8$  permil/amu,  $F(\text{Mg}) = 9.3 \pm 1.1$  permil/amu for AL-2,  $F(\text{Si}) = 0.43 \pm 1.3$  permil/amu,  $F(\text{Mg}) = 4.3 \pm 1.2$  permil/amu for AL-9, and  $F(\text{Si}) = 1.9 \pm 1.4$  permil/amu,  $F(\text{Mg}) = 6.4 \pm 1.6$  permil/amu for SH-1.

The inferred  $^{26}\text{Al}/^{27}\text{Al}$  ratios in melilites in these CAIs are nearly identical with the canonical value ( $5 \times 10^{-5}$ ) within the 2 sigma errors.

**Summary:** The Al-Mg ages of these CAIs suggest that the Mg isotopic fractionation was established when the CAIs initially formed, not by later metamorphic events. The average Si and Mg isotope fractionations for the three CAIs are positively correlated and agree well with those obtained by analyses of bulk CAIs [2], which are also consistent with the prediction of the model presented by [1]. The small Si isotope fractionation in AL-9 that seems to be akin to type A inclusions, compared with those in the other two typical type B CAIs, is also consistent with the expectation of [1] that type A CAIs should record a smaller amount of Si isotope fractionation for a given amount of Mg isotope fractionation. Thus, generally speaking, our results agree quite well with the

prediction of the model by [1] that type B CAIs experienced evaporation from a (nearly total) melt, which resulted in Rayleigh fractionation of Si and Mg isotopes.

**References:** [1] L. Grossman et al., (2000) *GCA*, 64, 2879-2894. [2] R. N. Clayton et al., (1988) *Phil. Trans. R. Soc. Lond.* A325, 483-501. [3] A.J.Fahey et al. (1987) *GCA* 51, 3215-3229. [4] J. N. Goswami et al. (1994) *GCA*, 58, 431-447. [5] A. M. Davis et al. (1991) *GCA*, 55, 621-637. [6] Hua X. et al. (2001) *MAPS* 36, A85. [7] Catanzaro E. J. et al. (1966) *J. Res. Nat. Bur. Stand.* 70a, 453-458. [8] Zinner E. (1989) *USGS B*, 1890, 145-162.