

ISOTOPIC AND ELEMENTAL FRACTIONATIONS PRODUCED DURING EVAPORATION OF THE ALLENDE CARBONACEOUS CHONDRITE. C. Floss¹, A. El Goresy¹, G. Kransel², W. Rammensee², H. Palme³ and E. Zinner⁴ ¹Max-Planck-Institut für Kernphysik, 69029 Heidelberg, Germany; ²Mineral.-Petrograph. Institut, Universität Köln, 50674 Köln, Germany; ³Max-Planck-Institut für Chemie, 55020 Mainz, Germany; ⁴McDonnell Center for the Space Sciences, Washington University, St. Louis, MO 63130.

Refractory inclusions (CAIs) from carbonaceous chondrites have provided important clues to the processes active in the early solar system. Some CAIs exhibit mass-fractionated isotopic (Mg, Si and Ca) compositions that deviate from normal values by much larger amounts than can be accounted for through igneous differentiation processes [1,2]; enrichments in both the heavy and light isotopes are observed, as expected for evaporation residues from a homogeneous melt and condensates from the concomitant vapor, respectively. In addition, large mass dependent fractionations of O (along with Mg and Si) are observed in F (Fractionation) and FUN (Fractionation and Unknown Nuclear) inclusions. Although these fractionations are commonly interpreted as the result of Rayleigh distillation processes in the solar nebula, the data, particularly correlated data on several elements, require multi-stage processes [1,2]. Some inclusions also show trace refractory element fractionations that are consistent with a distillation origin [3], although isotopic fractionations generally are not obviously related to either bulk or trace element compositions.

We have begun a series of evaporation experiments in order to better understand the processes and conditions under which mass-dependent isotopic and elemental fractionations are produced. We used powdered bulk Allende as a starting material. Individual samples, ranging in weight from 23 to 75 mg, were suspended from Re wire loops and evaporated from the liquid state to varying degrees in a vacuum furnace at 10^{-8} bar. Temperatures ranged from 1450 – 1750° C and run durations were from 15 to 180 minutes. Major element compositions of the residues were determined with the electron microprobe, and isotopic and trace element analyses were made with the ion microprobe. In these initial runs, mass losses ranging from 13–96 % were observed. Most residues consist primarily of olivine and glass with occasional tiny euhedral spinels. However, two of the samples with the highest mass losses ($\geq 95\%$) have lost more than 99% of their Fe, Mg and Si, and consist of a heterogeneous mixture of submicroscopic grains of Ca-aluminates. Individual analyses (which are probably not on discrete grains) give compositions that correspond to linear mixtures of $12\text{CaO}\cdot 7\text{Al}_2\text{O}_3$ and $\text{CaO}\cdot \text{Al}_2\text{O}_3$, and $\text{CaO}\cdot \text{Al}_2\text{O}_3$ and $\text{CaO}\cdot 2\text{Al}_2\text{O}_3$ (calcium dialuminate), respectively (Fig. 1). The Ca/Al ratios of these two residues are 7% and 51% lower than those of bulk Allende (0.68 and 0.37 versus 0.73), reflecting a preferential loss of Ca over Al in these samples. The evaporation behaviors of the major elements are consistent with the results of Wang *et al.* [4], who conducted similar experiments using a synthetic starting material of CI chondritic composition.

Isotopic measurements show normal Ca and Ti compositions for all residues analyzed so far, whereas both Mg and Si exhibit fractionations favoring the heavy isotopes (up to 45‰ per amu for Si), consistent with a Rayleigh distillation model. Oxygen isotopic compositions, measured in two of the residues, also show a mass-dependent enrichment of the heavy isotopes. Determination of appropriate evaporation reactions in a complex substance such as the one used here is difficult, as several different reactions are likely to have taken place at various stages during the evaporation process. Calculated gas/liquid partition coefficients (α 's) for Si evaporation are similar within errors to those observed by Davis *et al.* [5] for forsterite evaporation, for which the appropriate reaction is $\text{Mg}_2\text{SiO}_4(\text{l}) \rightarrow 2\text{Mg}(\text{g}) + \text{SiO}_2(\text{g}) + \text{O}_2(\text{g})$ [6].

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This implies that most Si evaporated as SiO_2 , although the slightly higher α values determined from our experiment suggest the involvement of heavier gas species or isotopic inhomogeneity during evaporation, as was also true for the Davis *et al.* experiment [5]. Partition coefficients calculated for Mg evaporation, however, are much closer to unity than those reported by these authors and, furthermore, are higher than theoretical α 's based on evaporation of Mg as either the monatomic species or as MgO, suggesting complex evaporation behavior for this element.

Trace element measurements in one of the residues with the highest mass losses show enrichments up to 45 x CI for most of the REE as well as for several other refractory lithophile elements (Fig. 2). Hafnium, expected to have an abundance similar to that of Zr, shows a 60% depletion relative to that element. Cerium and, to a lesser extent, Pr exhibit volatility-related depletions ($\text{Ce}/\text{Ce}^* = 0.002$; $\text{Pr}/\text{Pr}^* = 0.509$). Such depletions have been observed in several hibonite-rich inclusions [3] and in other laboratory evaporation experiments [3,4]. Wang *et al.* [4] noted that the oxidizing conditions implied by these depletions were probably due to the release of O during evaporation of major-element oxides. Despite their nominally similar volatilities under solar nebula conditions [7], Ba and Sr abundances differ, with the former being present at chondritic levels and the latter being enriched to 25 x CI. This is a further indication of oxidizing conditions during evaporation, because the volatility of Ba, relative to that of Sr, increases under oxidizing conditions [7]. Vanadium, which also becomes more volatile under oxidizing conditions, is present at less than chondritic abundances. Both elements are also depleted in HAL-type hibonite inclusions [3].

The results of these preliminary experiments are in good agreement with those of [4], despite the differences in starting materials, and show that isotopic fractionation effects as well as major and refractory trace element depletions observed in Ca-Al-rich inclusions can be duplicated through evaporation of chondritic material.

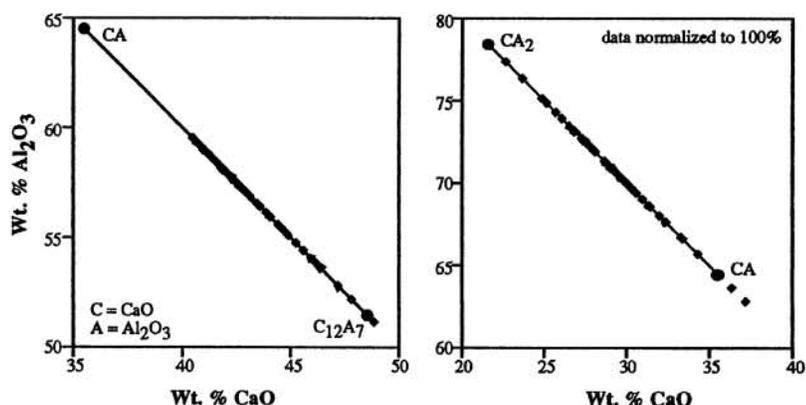


Figure 1

References: [1] Clayton *et al.*, (1985) In "Protostars and Planets II", 755-771. [2] Niederer and Papanastassiou (1984) *Geochim. Cosmochim. Acta* **48**, 1279-1293. [3] Ireland *et al.* (1992) *Geochim. Cosmochim. Acta* **56**, 2503-2520. [4] Wang *et al.* (1993) *Meteoritics* **28**, 454-455. [5] Davis *et al.* (1990) *Nature* **347**, 655-658. [6] Hashimoto (1990) *Nature* **347**, 53-55. [7] Davis *et al.* (1982) *Geochim. Cosmochim. Acta* **46**, 1627-1651.

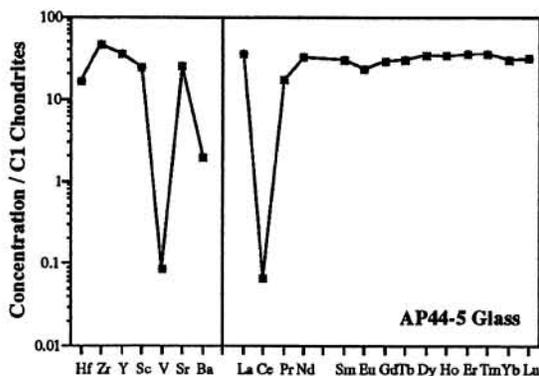


Figure 2