**SINGLE STAGE EVAPORATION OF SOLAR CONDENSATE DUST TO MAKE CAIS.** D. S. Ebel and L. Grossman<sup>1</sup>, Department of the Geophysical Sciences, 5734 South Ellis Ave., <sup>1</sup>also Enrico Fermi Institute, University of Chicago, Chicago, IL 60637 (debel@midway.uchicago.edu).

Introduction: In [1], we calculated the compositions of the residues which result from isothermal evaporation of CaO-MgO-Al<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub> (CMAS) liquids produced by melting the CMAS dust which condenses from solar gas at reasonable nebular conditions. The rate of evaporation was found to increase markedly with  $P(H_2)$ . The evaporation residues exhibit bulk compositions, and isotopic mass fractionations of Mg and Si, very similar to those observed in real Types A and B calcium-, aluminum-rich inclusions (CAI). Real CAIs, however, crystallized spinel and melilite during non-isothermal cooling from about 1700 or 1800 K, followed by Ca-pyroxene  $\pm$  anorthite nearer the solidus at ~1500 K. Subsequently, we investigated the conditions which produce normal and reverse zoning of melilite in CAIs evaporating non-isothermally [2]. Zoning records the balance between increased stability of Mg-, Si-rich melilite with decreasing temperature, and the evaporative loss of MgO and SiO<sub>2</sub> from the cooling liquid. The purpose here is to investigate the evolution of droplet chemistry during simultaneous cooling, evaporation, and crystallization of melted balls of dust previously condensed from a solar gas. We report cooling histories which result in solidified droplets having the bulk chemical compositions, isotopic compositions, and melilite composition zoning most consistent with those observed in real CAIs.

**Model:** The initial droplet contains CMAS oxides sufficient to make a liquid sphere of radius  $r_0=0.25$  cm (density 3.1 g/cm<sup>3</sup>), with oxides in the same proportions as the CMAS portion (excluding metal and TiO<sub>2</sub>) of the equilibrium solid assemblages which condense from solar gas at a total pressure (P<sup>tot</sup>) of 10<sup>-5</sup> bar [3]. Nearly all the Ca and Al have condensed once Si and Mg begin to condense on the trajectory shown in Fig. 1, so the Ca/Al mol ratio in these assemblages is solar [3]. The minerals in these assemblages are melilite + spinel (MgAl<sub>2</sub>O<sub>4</sub>) ± hibonite (CaAl<sub>12</sub>O<sub>19</sub>) ± Capyroxene. Representative precursor condensates ( $\alpha$ ,  $\beta$ ,  $\chi$ ,  $\delta$ , and  $\varepsilon$ ) were chosen as shown in Fig. 1.

The model assumes instantaneous heating to  $T_{max}$ , followed by linear cooling, all at a fixed pressure of pure H<sub>2</sub> gas. Only liquid evaporates, not solids, and solids are assumed not to affect evaporation by occluding the droplet surface. Diffusion in the liquid is assumed to be much faster than evaporation. Since pyroxene and anorthite crystallize late in real CAIs, the model only treats spinel and melilite. Based on diffusion coefficients in spinel [4] and melilite [5], melilite is assumed to be 100% fractionated, spinel not at all (0%). For each increment of melilite which crystal-

lizes, however, the volume of spinel present in the liquid which is replaced by the new melilite, is assumed to be trapped in the new melilite. At each temperature, the sequence of computations is: 1) equilibrium crystallization of spinel and melilite from the liquid [6], 2) evaporation of oxides from the coexisting liquid as in [1], 3) trapping of some spinel in the new melilite produced in step 1, 4) fractionation of melilite by addition of equilibrium melilite from step 1 to previously fractionated melilite, 5) recording of the droplet's final state, and 6) dissolution of spinel formed in step 1, but not trapped in step 3, back into the liquid; this spinel effectively re-equilibrates with the liquid in step 1.

Evaporation at each step is modeled using the Hertz-Knudsen equation. The evaporation coefficient for Mg,  $\alpha_{Mg}$  is a function of temperature (T, Kelvin):  $\log_{10}(\alpha_{Mg}) = -7787.34/T + 2.994$ , a fit to data of [7], and  $\alpha_{SiO} = 1.354 * \alpha_{Mg}$ , following [1]. Because SiO and Mg are the dominant gaseous species of Si and Mg,  $\alpha$  for other, insignificant, Si and Mg species are set to  $\alpha_{SiO}$  and  $\alpha_{Mg}$ , respectively. Negligible amounts of Al and Ca evaporate from CMAS liquids at T < 1800 K at P(H<sub>2</sub>) < 10<sup>-2</sup> bar. The effect of perfect kinetic isotope fractionation is calculated at each step using the Rayleigh equation, for Si, Mg, and O.

The melilite model addresses the solid solution series from gehlenite (Ge,  $Ca_2Al_2SiO_7$ ) to akermanite (Ak, Ca<sub>2</sub>MgSi<sub>2</sub>O<sub>7</sub>). The model used here differs from the one used in previous work [2, 3], which has been shown to be inconsistent with known phase relations, particularly for high Ge contents [8]. The new solution model, used with the liquid solution model of [6], greatly improves the agreement of predicted X<sub>Ak</sub> - T relations, and liquid compositions, with experimental results of [9] and [10], and with the Ge-Ak phase diagram [11]. For experimental liquid-melilite pairs with  $X_{Ak} < 0.1$ , the predicted T of melilite appearance (T<sub>mel</sub>) is in excellent agreement, but the predicted  $X_{Ak}$  exceed those observed by 0.04-0.06. Compared to experiments with 0.15<X<sub>Ak</sub><0.20, T<sub>mel</sub> is high by ~30 K, and X<sub>Ak</sub> low by 0.02-0.04. The new model reproduces the phase diagram very closely for  $X_{Ak} < 0.7$ .

**Results:** Real CAIs result from a combination of solar nebular processes. Important measured properties which record those processes are CAI bulk chemical compositions, degree of bulk isotopic fractionation of Mg and Si, and chemical zonation of melilite grains, which become Ak-rich from core (initial  $X_{Ak}$ ) to rim in nearly all CAIs (normal zoning). It has been deduced from textural criteria [12] that the maximum T ( $T_{max}$ ) reached by composition CAIB (Fig. 1) was ~1675 K,

about 20 K above  $T_{mel}$ . The B2s are closest to CAIB in composition. In B1s, melilite appears to have crystallized from the liquid [13], therefore we assume  $T_{max}>T_{mel}$ . Experimental work on a CTA-like composition [14, #98] showed that  $T_{mel}$  is ~20 K below the liquidus, and did not yield CTA melilite textures at superliquidus  $T_{max}$ . That work, combined with little evidence for relict melilite in CTAs [15], implies that  $T_{liquidus}>T_{max}>T_{mel}$ . Because CAIs contain progressively more MgO and SiO<sub>2</sub> from Type A to B1 to B2,  $T_{mel}$  decline in the same order, so the constraint  $T_{max}>T_{mel}$  results in progressively lower  $T_{max}$  (see Fig.1, Table 1).



One possible type A precursor ( $\beta$  in Fig. 1) evolves into the composition field of real Type As from a very high T<sub>max</sub>, 1793 K. The evaporation rate is so high at P(H<sub>2</sub>)=10<sup>-3</sup> bar that melilite is reversely zoned, even at a cooling rate of 60 K/hr (not shown in Figs). At the same T<sub>max</sub>, but a lower P(H<sub>2</sub>) of 10<sup>-6</sup> bar, and cooling rates of -5 and -3 K/hr, this precursor yields bulk compositions (Fig. 1), isotope fractionations (Table 1), and normally zoned melilites (Fig. 2), all consistent with those observed in CTAs. For other precursor compositions, below, we report only results at P(H<sub>2</sub>)=10<sup>-6</sup> bar.

In more SiO<sub>2</sub>-poor compositions, such as  $\alpha$ , so much melilite crystallizes at very high T that it re-

places most of the liquid in a small T interval. A higher  $T_{max}$  or slower cooling rate allows evaporation to produce greater change in bulk composition, both above and below  $T_{mel}$ . When  $\alpha$  is evaporated from 1813 K at -5 K/hr, melilite replaces 90 volume % of the liquid by 1770 K, so most of the resulting melilite is normally zoned (Fig. 2), and the bulk composition evolves through the Type A field, ending at slightly more MgO-poor compositions.

Composition  $\chi$  is a possible precursor for a range of Type B1 CAIs, if it cooled from  $T_{max}$ =1753 K, at -1 or -2 K/hr, at P(H<sub>2</sub>)=10<sup>-6</sup> bar. These cooling rates produce  $F_{Mg}$  and  $F_{Si}$  in the middle of the ranges reported for type Bs [1, fig. 8]. More SiO<sub>2</sub>-rich B2s require more SiO<sub>2</sub>-rich precursors, but melilite crystallizes from compositions such as  $\delta$  and  $\epsilon$  at much lower T than from more SiO<sub>2</sub>-poor compositions. For precursor  $\delta$  to evaporate into the B2 field, crystallize normally zoned melilite within 20 K of  $T_{max}$ , yet not exceed the  $F_{Mg}$  observed in B2s,  $T_{max}$  must be less than 1723 K, and the cooling rate must be  $\leq 1$  K/hr, at P(H<sub>2</sub>)=10<sup>-6</sup> b.

For precursor  $\varepsilon$  the difficulty is greater. Cooled from  $T_{max}$ =1673 K at -5 K/hr,  $T_{mel}$ =1620 K, ini $X_{Ak}$ =0.28. Cooled at -3 K/hr,  $T_{mel}$ =1640 K, ini $X_{Ak}$ =0.25, but melilite is reversely zoned from core to rim. Unless the constraint  $T_{max}$ - $T_{mel} \leq 20$  K is relaxed, the more SiO<sub>2</sub>-rich B2s cannot have resulted from single-stage evaporation of precursor  $\varepsilon$ . It may be that the trajectories of real condensate bulk compositions are steeper than the one shown in Fig. 1. Constraints may be met with a wider range of thermal histories, if ambient gases contain Mg and Si.

<u>Table 1</u> : Results, trajectories of Fig. 1, $P(H_2)=10^{-6}$ bar.												
	<u>T</u> <sub>max</sub>	K/hr	T <sub>mel</sub>	$iniX_{Ak}$	<u>F</u> <sub>Si</sub>	<u>F</u> <sub>Mg</sub>	<u>F</u> o					
α	1813	-10	1800	6.31	0.46	4.29	0.65					
α	1813	-5	1801	5.91	0.62	6.23	1.03					
β	1793	-5	1774	12.3	0.64	4.84	1.01					
β	1793	-3	1777	11.7	1.21	7.84	1.67					

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δ	1693	-0.5	1674	24.3	1.75	6.11	2.07
δ	1693	-1.0	1669	25.3	0.83	3.00	1.03
χ	1753	-1	1741	16.8	2.37	10.18	2.74
χ	1753	-2	1738	17.5	1.05	5.13	1.37

**References:** [1] Grossman L *et al.* (2000) GCA **64**, 2879. [2] Ebel DS and Grossman L (2000) MaPS Suppl. **35**, A49. [3] Ebel DS and Grossman L (2000) GCA **64**, 339. [4] Sheng YJ *et al.* (1992) GCA **56**, 2535. [5] Morioka H and Nagasawa M (1991) GCA **55**, 751. [6] Berman RG (1983) PhD, UBC. [7] Richter *et al.*, GCA, submitted. [8] Yoneda S and Grossman L (1995) GCA **59**, 3413. [9] Beckett JR (1986) PhD, Chicago. [10] Stolper E (1982) GCA **46**, 2159. [11] Osborn EF and Schairer JF (1941) Am J Sci **239**, 715. [12] Stolper E and Paque J (1986) GCA **50**: 1785. [13] MacPherson GJ and Grossman L (1981) EPSL **52**: 16. [14] Paque JM and Stolper E (1984) LPS **XV**, 631. [15] Simon SB *et al.* (1999) GCA **63**: 1233.