

COARSE-GRAINED REFRACTORY INCLUSIONS: CONDENSATES, EVAPORATION RESIDUES, OR BOTH? EVIDENCE FROM MAJOR ELEMENT BULK COMPOSITIONS. S. B. Simon, D. S. Ebel, L. Grossman¹ and A. M. Davis¹. Dept. Geophys. Sci., 5734 S. Ellis Ave, Univ. of Chicago, Chicago, IL 60637. ¹Also Enrico Fermi Inst., Univ. of Chicago. (sbs8@midway.uchicago.edu)

Introduction. It has long been known that, but not understood why, bulk compositions of coarse-grained refractory inclusions (CAIs) in CV3 chondrites generally differ from bulk equilibrium condensates calculated for a gas of solar composition. Literature data for major element oxide compositions of most Types A and B inclusions may be in error, however, due to non-representative sampling of spinel relative to other phases, for example, because of small sample sizes and the heterogeneous distribution of spinel within CAIs. We corrected reported compositions to the solar CaO/Al₂O₃ ratio by addition or subtraction of spinel, and here we compare the revised CAI compositions to those expected from equilibrium condensation. We also performed thermodynamic calculations to evaluate the degree to which open-system evaporation may have affected CAI bulk compositions.

Sampling and Analysis Techniques. Refractory inclusions in Allende and other members of the oxidized subgroup of CV3 chondrites underwent extensive, relatively low-temperature secondary alteration, which introduced volatile elements and may have also preferentially removed some refractory elements relative to others. In this paper, we used only lightly altered inclusions from the reduced subgroup of CV3 chondrites and those from Allende whose major element compositions were obtained by techniques designed to avoid or correct for effects of secondary alteration. Inclusions selected for this work are all coarse-grained Types A and B inclusions that are not hibonite-rich analyzed by [1- 9], excluding one from [8] which contains abundant alteration products. All analyses were re-normalized to 100 wt % CaO+MgO+Al₂O₃+SiO₂ (CMAS), thus removing 0.5-1.5 wt % TiO₂ and all volatile elements from further consideration.

Comparison with Condensation Calculations. The data were compared to three equilibrium bulk condensate trajectories calculated using the method of [10] for conditions selected to illustrate a range of condensate assemblages: a gas of solar composition at $P^{\text{tot}} = 10^{-5}$ bar, where condensation is entirely to crystalline phases; a solar gas at $P^{\text{tot}} = 10^{-1}$ bar, where partially molten CaO-, Al₂O₃-rich assemblages condense; and at $P^{\text{tot}} = 10^{-3}$ bar in a system enriched in C1 dust relative to gas by a factor of 20 compared to solar composition, in which CMAS partial melts also condense [11]. Despite these differences, the three bulk condensate trajectories lie very close to one another. On some oxide-oxide plots, the inclusions plot along the equilibrium condensation path, but on most plots the data scatter widely, with many inclusions lying far from the predicted trends. Particularly egregious is the deviation of the data from the condensation path ($P^{\text{tot}} = 10^{-5}$ bar) on the CaO-Al₂O₃ plot, Fig. 1. Since all inclusions selected for this work are hibonite-poor, it can be assumed that they formed below the temperature at which both Al and Ca are totally condensed [11]. All hibonite-poor CAIs should, therefore, have the solar CaO/Al₂O₃ ratio and lie along the left arm of the trajectory shown on Fig. 1. Instead, very few of the inclusions lie along that line, and they actually form a trend which cross-cuts it, with a range of CaO/Al₂O₃ ratios from 34%

below the chondritic value to over 100% above it. Another puzzling result is the distribution of data (open symbols) on the MgO-Al₂O₃ plot, Fig. 2. Because fassaite is predicted to form during condensation by reaction of gaseous Mg and Si with melilite, Type Bs, which are fassaite-rich, are expected to have higher MgO contents than Type As, which are melilite-rich and fassaite-poor. Although this is observed, the addition of MgO and SiO₂ to make fassaite must also dilute the Al₂O₃ contents, and this is not observed.

On most plots, the composition of spinel lies on the opposite side of the condensation trends from the compositions of the melilite components. This suggests that inclusion compositions that originally lay on the condensation trends could have been artificially dispersed about the trends by sampling non-representative proportions of spinel relative to melilite. We assumed that the only reason measured inclusion compositions deviate from some of the calculated composition trajectories is that the samples taken for analysis contain more or less spinel relative to other phases compared to the actual compositions. To correct for this, we performed a calculation in which sufficient Al in the form of MgAl₂O₄ was either added to or subtracted from the bulk chemical composition of each inclusion such that the corrected composition has the cosmic CaO/Al₂O₃ ratio. This resulted in a dramatic reduction in scatter of the data, and yielded trends that match the condensation trajectories very closely. There is vast improvement on the CaO-SiO₂ plot, which is impressive because spinel contains neither of these oxides. Evidence that the corrected analyses are more accurate than the raw data also comes from the improved consistency between chemical analyses and mineralogical compositions. For example, most Type B inclusions now have lower Al₂O₃ contents, consistent with their higher MgO (Fig. 2) and SiO₂ contents, than Type As, as is expected for inclusions in which much melilite has been converted to fassaite. There is much overlap between bulk compositions of Types B1 and B2 inclusions, but we note that B1s tend to have lower SiO₂ (Fig. 3) and higher CaO and Al₂O₃ contents than B2s, leading to higher calculated gehlenite saturation temperatures for B1s than B2s.

The plots involving MgO show the greatest deviations of the data points from possible condensation paths. On the SiO₂-MgO plot (Fig. 3), the corrected data for Type B inclusions scatter relative to one another and deviate considerably from the condensation trajectory. One interpretation of this is that the inclusions were affected by an additional process after condensation, one that preferentially changed the original MgO and SiO₂ concentrations. These are the two most volatile oxides considered here, and most prone to evaporation. Had any condensate assemblage whose composition lies on any of the condensation paths been heated so that it underwent equilibrium evaporation in a gas having the same composition and P^{tot} as the one it had condensed from, however, its composition would have merely evolved along the same composition trend, but in the up-temperature direction. If, however, a condensate assemblage were heated in a medium different from the one from which it had condensed, in particular, one not saturated in condensable elements, the

evolution of its composition during evaporation would not be restricted to the condensation path.

Effect of Evaporation on Condensate Chemical Compositions. We now address the question of whether the evaporation of MgO and SiO₂ from predicted high-temperature condensates can bring their bulk chemical compositions into even closer agreement with those of coarse-grained inclusions. Open-system evaporation of partially molten assemblages was modelled [12] at $P(H_2) = 1 \times 10^{-8}$ bar at 1700, 1800, 1900 and 2000K for various starting compositions along the condensate trajectory. At 2000K, SiO₂ contents decrease so steeply as evaporation proceeds that at the correct MgO contents, predicted SiO₂ contents are below those of most Type B inclusions. The slopes of the evaporation paths gradually become flatter, however, with progressively lower evaporation temperatures. As a result, at 1700K (Fig. 3), the predicted field of evaporation residues overlaps the bulk compositions of almost all Type B inclusions. Most Type A compositions are probably within measurement error of the condensate trajectory. Bulk chemical changes due to evaporation of large fractional amounts of MgO and SiO₂ from Type A compositions are relatively small because of their low MgO and SiO₂ contents, and the absolute change in SiO₂ content for a particular fraction of the Mg evaporated is small for these SiO₂-poor assemblages. Bulk chemical compositions of many Type A and B CAIs are compatible with up to ~30 % evaporation of Mg and less than 10% of Si at 1700K. These estimates are within the ranges inferred from isotopic compositions of CAIs [12].

Conclusions. Coarse-grained CAIs are commonly sampled unrepresentatively, especially with respect to spinel. Correcting bulk analyses of hibonite-poor inclusions to the solar CaO/Al₂O₃ ratio by addition or subtraction of MgAl₂O₄ reduces the scatter of the data and results in trends that fall along paths of bulk equilibrium condensates. Much of the remaining scatter and deviation from condensate trends, especially on the MgO-SiO₂ plot, can be accounted for by open-system evaporation of partially molten equilibrium condensates. In a companion abstract [12], we show that the degrees of evaporation inferred from major element oxide compositions are within the ranges inferred from Mg and Si isotopic compositions of refractory inclusions; however, the major element data are from one suite of refractory inclusions and the isotopic data from another. The ultimate test of the model awaits measurement of the bulk chemical compositions of whole inclusions and the magnesium, silicon and oxygen isotopic compositions of the same inclusions.

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