EVAPORATION OF CaO-MgO-Al₂O₃-SiO₂ MELTS:
EXPERIMENTS AND THERMODYNAMIC MODELING.
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Introduction: The coarse-grained texture of igneous Ca-, Al-rich Inclusions (CAIs) indicate that their precursors have experienced a high-temperature melting event followed by slow cooling. The melting under low-pressure solar nebula conditions could have resulted in chemical and isotopic fractionations which are now recorded in CAIs. To understand the evaporation kinetics and thus to place quantitative constraints on conditions at which the CAIs were formed, a large number of laboratory experiments were conducted in which CaO-MgO-Al₂O₃-SiO₂ (CMAS) melts were evaporated in a vacuum at 1600° to 1900°C (e.g., [1–3] and references therein). These experiments successfully reproduced the major chemical and isotopic features of CAIs. The evaporation process could also be modeled by using a thermodynamic approach as it was applied in [4–6], for example. Here we compare experimental results on evaporation of two CAI-like melts with the ones calculated using the stoichiometric-Margules solution model for CMAS system [7] and using the ideal associated-solution model [8].

Results and Discussion: Experiments on evaporation of Type B CAI-like (CAIB) melt [1] show that evaporation starts with a much faster loss of Si over Mg followed by their evaporation at about the same rates as evaporation proceeds. The experiments also showed that despite a strong temperature effect on evaporation kinetics of Mg and Si (evaporation rates of Mg are ~2×10⁻⁹, ~1×10⁻⁸, (2–6)×10⁻⁸ and (2–3)×10⁻⁷ moles/cm²/s at 1600°, 1700°, 1800° and 1900°C, respectively), the temperature had only a small effect on evaporation trajectories.

The evaporation of CAI-like melts could also be modeled thermodynamically using an approach of [4] in which the equilibrium gas composition above a melt was calculated first, then the amount of Ca, Mg, Al and Si in the gas phase was subtracted from their amounts in the starting melt, and the resulting new composition melt was considered as a new starting material and the procedure continued.

Such an approach using Berman’s CMAS model [7] was used in [1] to model evaporation of CAIB melt. The calculations have reproduced the major chemical features observed in experiments, but have shown that temperature would strongly effect the evaporation trajectory which contradicts the experimental observations. The calculated trends can match the experimental ones if evaporation coefficients of Mg (γ₉₉) and Si (γ₅₈) of the model vary such that γ₉₉/γ₅₈ should be ~1 to match the experiments at 1600° and ~0.5 at 1900°C. In other words, the evaporation coefficients during evaporation of the same composition melt should strongly depend on temperature. In addition, in order to match the experimental data on evaporation of CMS-1 FUN CAI-like (CMS-1) melt that was evaporated at 1900°C [3], the γ₉₉/γ₅₈ is ~2 is required, which is very different from γ₉₉/γ₅₈ ~0.5 for CAIB melt at the same temperature. The reason why γ₉₉ and γ₅₈ should strongly depend on the temperature and melt composition is unclear.

To model evaporation of CAIB and CMS-1 melt here, we used a thermodynamic model [9] in a framework of the ideal associated-solution theory [8]. The major advantage of such a model is that it utilizes activities of components in high-temperature melts directly measured by Knudsen effusion mass spectrometry (KEMS) rather than being obtained indirectly. The model also allows calculating thermodynamic properties of a melt within a wide range of composition in the system of interest using available data for the most important melt components. Using such a model and thermodynamic properties of melt components determined by KEMS at 1300 to 2200°C, we calculated evolution of chemical composition and activities of components for CAIB and CMS-1 melts. It was found that in agreement with the experimental results, the calculated trends for CAIB melt shows very weak temperature dependence of the evaporation trajectory and fit the experimental ones quite nicely without changing evaporation coefficients of Mg and Si. It should be mentioned that in these calculations we used evaporation coefficients equal to 1, which are typical for molten oxides (see [10] for experiments on molten SiO₂ and [11] on molten Al₂O₃), as well as for liquids with spherically symmetrical molecules and atomic liquids [12].