Thermodynamics and kinetics of evaporation of CMAS melts

R. A. MENDYBAEV[1] AND S. I. SHORNIKOV[2]

[1]University of Chicago, Chicago, IL 60637, USA (ramendyb@uchicago.edu)
[2]V.I. Vernadsky Institute of Geochemistry and Analytical Chemistry, Moscow 119991, Russia (sergey.shornikov@gmail.com)

Majority of Ca-,Al-rich inclusions (CAIs) in chondrites have experienced high-temperature melting followed by slow cooling as indicated by their coarse-grained igneous textures. The melting and evaporation under low-pressure conditions result in chemical and mass-dependent isotopic fractionations as recorded in CAIs. The effects have been successfully reproduced in vacuum and in low-pressure H_2 laboratory evaporation experiments [1, 2].

Several attemps have been made to model the evaporation thermodynamically [3–5] using Berman's CMAS model or its combination with the MELTS. The evaporation process can be described by the Hertz-Knudsen equation and for the CMAS system the ratio of the evaporative fluxes of Mg and Si is proportional to their evaporation coefficients (γ_i) and activities (a_i) in the melt. The calculated a_i in several melts using different available models (FactSage, Thermocalc, CMAS) showed variations in a_{CaO}, a_{MgO} and a_{Al2O3} up to a factor of ~10 at 2173 K which results in very different evaporation trends. We have calculated the evaporation trajectories for a number of melt compositions (melilitic, anorthitic, forsteritic) using model of [6] which showed very good agreement with the experimental data. For example, in agreement with the experiments the calculated trajectories for melilitic melts show very weak temperature dependence and did not require adjusting γ_{Mg} and γ_{Si} to fit the experimental trends (γ_{Mg}=γ_{Si}=1 was used in our calculations) unlike CMAS based calculations. The model is based on the theory of ideal associated solutions, which consider a solution as an ideal mixture of monomeric molecules and associative complexes at all concentrations. The major advantage of the model is that it utilizes a_i directly measured in Knudsen cell experiments at 1600–2500 K rather than being obtained indirectly. We will expand the experimental database on evaporation kinetics (to CMAS + TiO_2 system first) to further check correctness of the model for complex systems.

[1] Richter et al (2007) *GCA* **71**, 5544–5564. [2] Mendybaev et al. (2013) *GCA* **123**, 368–384. [3] Grossman et al. (2000) *GCA* **64**, 2879–2894. [4] Alexander et al. (2002) *MAPS* **37**, 245–256. [5] Ebel (2005) *GCA* **69**, 3183–3193. [6] Shornikov (2009) *Heralds Earth Sci. Rus. Acad. Sci.* **27**, 1–3.