Experimental quantification of Na solubility in silicate melts. Application to chondrule formation

R. Mathieu[1], G. Libourel[1,2], E. Deloule[1], L. Tisssandier[1], [1]CRPG-Nancy Université-INSU-CNRS, UPR2300, BP 20, 54501 Vandoeuvre-lès-Nancy, France. [2]ENSG-Nancy Université, 54500 Vandoeuvre-lès-Nancy, France (Email: rmathieu@crpg.cnrs-nancy.fr)

Introduction

Alkalies are among the most variable elements in chondrule mesostases (e.g. from 0 up to ~ 15wt% for Na_2O) [1, 2]. Various scenarii have been proposed to explain this scattering of compositions, such as various degrees of evaporation, direct condensation from gaseous nebular environments [3] or low temperature alteration in the parent body [4].... The impossibility to clearly choose one of these models comes, at least in part, from the poor knowledge of alkali behavior in silicate melts and gaseous environments.

It has now been well established that alkali evaporation rates are clearly dependent on temperature and fO_2 conditions, but, till now, no clear thermodynamic model allows us to predict what will be, e.g. the solubility of these elements in silicate melts or their partition coefficients between these melts and their gaseous environment.

To tackle this issue, we have developed a new device to control alkali-metal oxide activity in molten silicates (aNa_2O) by equilibration of melts with a gaseous environment of known Na and O partial pressures (P_{Na} and P_{O_2}).

Experimental design

Our method [5] consists in equilibrating at high temperature, molten silicates with alkali vapor produced by a reference Na_2O-$xSiO_2$ ($2 \leq x \leq 6$) binary melt with a known aNa_2O. Sodium evaporates from the source according to eq. 1:

$$Na_2O^{(source)} \rightarrow 2Na_{(g)} + 1/2\ O_2 \quad (1)$$

Na(g) can then condense in (partially) molten samples following eq. 2:

$$2Na_{(g)} + 1/2\ O_2 \rightarrow Na_2O^{(sample)} \quad (2)$$

Once the equilibrium is reached [5], alkali oxide activity in samples is directly fixed by the source according to eq. 3:

$$aNa_2O_{(sample)} = aNa_2O_{(source)} \quad (3)$$

Results and Discussion

Na solubility has been investigated in a very wide range of compositions belonging to the CaO-MgO-Al_2O_3-SiO_2 system (165 samples with $0 < CaO$ and $MgO < 40$; $0 < Al_2O_3 < 45$; $0 < SiO_2 < 100$; in wt%, and NBO/T varying from 0 to 3). Different PNa (from 10^{-4} atm to 10^{-6} atm) have been imposed by varying Na_2O-$xSiO_2$ ($2<x<6$) sources [6,7,8].

At equilibrium, thermochemical variables such as sodium solubility S_{Na} or activity coefficients of Na in melts $\gamma Na_2O^{(sample)}$ can be obtained. The influence of sample composition on these parameters is then tested, with a very important role of melt polymerization and Ca/Mg ratio, and can be modelized with quite simple equations.

Moreover, Na entering effects on phase relationships have been characterized in the samples and this has lead to the development of a new generation of phase diagrams.

When applying these results to chondrule formation, it is possible to constrain the gaseous environment when these objects were (partially) molten, i.e. during the first steps of formation of the Solar System.

References

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