THERMODYNAMICS OF EVAPORATION OF CHONDRULE MELTS OF SARATOV CHONDRITE.

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The formation of chondrule chondrites is given increased attention by researchers. Despite the success of modern experimental studies, the conditions for the formation and evaporation of chondrule melts remain unclear [1]. It is known that K in elementary, oxide, chloride and other simple forms is more volatile than Na, that is also observed in experiments on basalt and more basic melt evaporation [2]. Other authors suggest a high volatility of sodium [1].

In this regard, the results of a single mass spectrometric experiment [3] on the evaporation of pyroxene chondrules of Saratov chondrite from a tungsten cell (according to Knudsen) at 1300–2400 K are of particular interest. The chondrules of Saratov chondrite belong to type IIB and are an oxide material (wt %): SiO₂ (54.44), TiO₂ (0.13), Al₂O₃ (3.20), FeO (13.17), MgO (24.98), CaO (2.24), Na₂O (1.44) and K₂O (0.40). A relatively low melting point and insignificant fractionation are typical for these chondrules. The details of the evaporation experiments are described by Yakovlev et al. [4]. The composition of the residual melt (Figs. 1a and 2) was calculated from the partial pressures (*p*) of the vapor species over the chondrule melt *vs*. the temperature and evaporation time [3].

As follows from Fig. 1a and 2, the evaporation process of chondrules can be divided into two stages. The first stage is in the temperature range of $1300-1600 \, \text{K}$ and is characterized by the evaporation of K_2O and Na_2O from the melt with an almost unchanged content of the remaining oxides (Fig. 1a). In this case, the melt loses approximately 2 wt % during evaporation, which corresponds to the total amount of evaporated alkaline oxides. Note that the symbateness of the change in the K_2O and Na_2O contents during evaporation indicates an approximate equality of their volatility.

The second stage is in the temperature range 1600-2400 K, where the remaining oxides evaporate in the order of their individual volatility: FeO, SiO₂, MgO, TiO₂, CaO and Al₂O₃ (Fig. 2). At this evaporation stage an inversion occurs in the MgO / SiO₂ content ratio in the chondrule melt, which is due to the proximity of their volatilities. A decrease in the FeO, MgO and SiO₂ content in the melt during evaporation leads, respectively, to a symbatic increase in the TiO₂, CaO and Al₂O₃ content up to 5 wt % of residual melt. Then the melt successively loses TiO₂ and CaO.

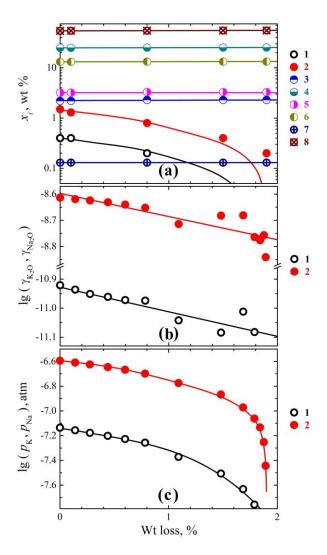


Fig. 1. The chondrule residual melt composition of Saratov chondrite during evaporation at 1406-1569 K (a), the K_2O and Na_2O activity coefficients (b) and the K and Na partial pressures over the melt at 1450 K (c). Table of symbols:

- (a) the results of [3] shows the symbols: K_2O (1), Na_2O (2), CaO (3), MgO (4), Al_2O_3 (5), FeO (6), TiO_2 (7), SiO_2 (8); the lines correspond to the calculated results; (b) the calculated K_2O (1) and Na_2O (2) activity coefficients;
- (c) the calculated K (1) and Na (2) partial pressures.

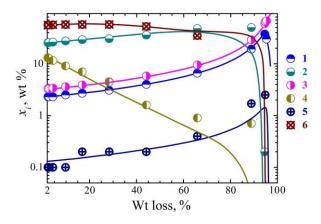


Fig. 2 The chondrule residual melt composition of Saratov chondrite during evaporation at $1569-2147~\rm K$. The results of [3] shows symbols: CaO (1), MgO (2), Al₂O₃ (3), FeO (4), TiO₂ (5), SiO₂ (6); the lines correspond to the calculated results.

The theoretical calculations of thermodynamic properties of the chondrule melt of Saratov chondrite at 1300-2400~K were performed using a semi-empirical model designed to calculate activity coefficients in the CaO-MgO-Al₂O₃-FeO-TiO₂-SiO₂ melts in a wide range of compositions and temperatures [5].

The initial information on the condensed phases and vapor species of the gas phase over the CaO–MgO–Al₂O₃–FeO–TiO₂–SiO₂ melts was supplemented with thermodynamic data for the K₂O–SiO₂ [6] and Na₂O–SiO₂ [7] melts, as well as K₂O–Na₂O–SiO₂ melts [8].

It can be seen from Figs. 1a and 2 that the calculated composition of the chondrule residual melts of Saratov chondrite during evaporation corresponds to the experimental data [3]. The observed slight deviations in the Na₂O (Fig. 1a), as well as FeO and MgO (Fig. 2) content, may be due to the low content of Na₂O in the residual melt (0.1 wt %), or an increase in experimental errors at the noticeable decrease in the residual melt amount (up to 10 wt %).

Evaporation of alkaline oxides from the chondrule melt is accompanied by symbatic linear logarithmic dependences of their activity coefficients (Fig. 1b), which are quite low $(10^{-12}-10^{-8})$. Note that the evaporation of these oxides ends simultaneously (Fig. 1c). Despite the K₂O high individual volatility, its partial pressure is lower than the Na₂O partial pressure due to its low content in the melt. In addition, the volatility is determined by the component activity coefficient, depending on the acidity (basicity) of the melt. The chondrules of Saratov chondrite contain a significant amount of SiO₂ (54.44 wt %), which provides a high acidity of the

melt, which does not allow K₂O to exhibit high volatility during evaporation.

An interesting feature of the chondrule melt evaporation is the correlation of the CaO and SiO₂ contents in the melt and the atomic and molecular oxygen content in the gas phase over the melt. Fig. 3 shows that the main source of atomic oxygen is CaO, which represents the melt basicity, and the main source of molecular oxygen is SiO₂, which characterizes the melt acidity.

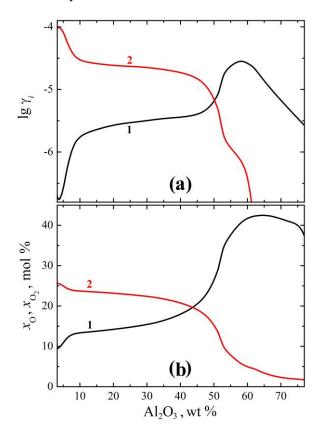


Fig. 3. The CaO (1) and SiO_2 (2) activity coefficients (a) and the atomic (1) and molecular (2) oxygen content (b) in the gas phase over the chondrule residual melts of Saratov chondrite vs. the Al_2O_3 content in the melt at 2000 K.

References: [1] Ebel D. et al. (2018) Chondrules. Records of protoplanetary disk processes, 151–174. [2] Yakovlev et al. (1972) Dokl. AN SSSR, 206, 970–973 [3] Yakovlev et al. (1987) Meteoritika, 46, 104–118. [4] Yakovlev et al. (1984) Meteoritika, 43, 126–132. [5] Shornikov S. I. (2019) Geochem. Int., 57, 865–872. [6] Shornikov S. I. (2013) Experiment in Geosciences, 19, 108–111. [7] Shornikov S. I. (2014) Ibid, 20, 48–51. [8] Shornikov S. I. and Yakovlev O. I. (2020) Physico-chemical and petrophysical research in Earth sciences, 22, 293–296.