



Solution behavior of reduced N-C-H-O volatiles in FeO-Na₂O-SiO₂-Al₂O₃ melt equilibrated with molten Fe alloy at high pressure and temperature

A. Kadik (1) and Y. Litvin (2)

(1) V.I. Vernadsky Institute of Geochemistry and Analytical Chemistry, RAS, Kosygin St. 19, Moscow 119991, Russia (kadik@geokhi.ru, +7 (495) 938 2054), (2) Institute of Experimental Mineralogy, RAS, Chernogolovka, Moscow distr. 142432, Russia kadik@geokhi.ru / 7-495-137-7200

In a series of experiments conducted at 1.5 GPa, 1400°C and 4 GPa, 1550°C, and fO_2 from 2 to 4 log units below IW buffer we have characterized the nature and quantified the abundance of N, C and H species dissolved in a model silicate melt (FeO + Na₂O + Al₂O₃ + SiO₂) in a manner similar to that reported by [1, 2]. To elucidate the mechanisms of nitrogen and hydrogen dissolution in magmas, we studied the glasses produced by quenching the melts using infrared and Raman spectroscopies. Experiments indicate that under the reduced conditions corresponding to the fO_2 path during metal segregation and self-oxidized of mantle [3] and magma ocean [e.g. 4, 5] the silicate melt would contain species with N-H bonds (NH₃, NH⁴⁺, NH²⁻, NH²⁺) as well as N₂, C-H bonds (CH₄), H₂ and oxidized H species (OH⁻ and H₂O). The formation of N-H bonds in the reduced silicate melts results in a increase in nitrogen solubility that can reach 1–2 wt. %. It is suggested that significant amounts of nitrogen, comparable to those estimated for the present-day mantle, could have been incorporated in the early Earth by dissolution in reduced magma ocean. Experimental data testify to a strong influence of pressure, fO_2 , and hydrogen on distribution of nitrogen between metallic and silicate melts, (DN_{met/sil}). It is found that at 1.5 GPa, 1400°C, $\Delta \log fO_2 = -3.7$ and 4 GPa, 1550°C, $\Delta \log fO_2 = -3.3$ the values of DN_{met/sil} are equal to 0.48 and 0.43 accordingly. The experimental studies at a pressure corresponding to a depth of 100–150 km have shown that the self-oxidation of magma ocean with fO_2 increasing from 4 to 2 log units below fO_2 (IW) [3,4,5] is characterized by a decrease in the amount of species with N-H and C-H bonds and an increase in the content of oxidized hydrogen species, such as OH⁻ and H₂O. These dissolution features make fO_2 a critical factor in the formation of N-O-H volatile species during the large-scale melting of the early Earth.

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[2] Kadik et al (2011) Geochem. Inter. 49 (5), 429–438;

[3] Galimov (2005) EPSL 233, 263-276, [4] Wood et al (2006) Nature 441, 825-833, [5] Frost et al (1008) Phil Trans Royal Soc A 366, 4315–437.

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