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Role of hydrogen and oxygen fugacity on incorporation of nitrogen and carbon in reduced magmas of the early Earth

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In a series of experiments in the system Fe-bearing melt + molten Fe metallic phase + N+ H conducted at 4 GPa and 1550°C and logfO2 from 2 to 4 below logfO2 (IW), we have characterised the nature and quantified the abundance's of N- and H-compound dissolved in an model silicate melt (NaAlSi3O8 80% wt +FeO 20% wt). Experiments were carried out in an anvil-with-hole apparatus. The technique of fO2 buffering employed relies upon the diffusion of H2 through Pt to achieve equal chemical potentials of H2 in the inner Pt capsule and outer solid fO2 buffer assembly in the presence of H2O. The fO2 imposed on the charge is controlled by the equilibrium between H2 buffered externally, and the Fe-bearing melt. To create a low fO2 in the experiments, 2, 3, 5 and 7 wt % of finely dispersed Si3N4 was added to the glass powder (NaAlSi3O8 80% + FeO 20% wt). The initial Si3N4 was unstable under experimental conditions and was completely consumed according to the reactions: Si3N4(initial) + 3O2 $\rightarrow$ 3SiO2(melt) + 4N(melt) with the subsequent participation of nitrogen in reactions with hydrogen and components of silicate melts. The infrared and Raman spectroscopy of glasses indicates a remarkable feature of N-H interaction with a reduced silicate: an appreciable change in the mechanism of their dissolution with a decrease in fO2. The most part of nitrogen reacts with hydrogen with formation of N-H complexes. The most likely nitrogen-bearing species is represented by NH3 and NH4+ group. Except for N-H complexes hydrogen is expressed under the oxidized form OH and H2O. Some hydrogen is present in a melt in the molecular form. The amount of H and N dissolved in the glasses was measured by ion microprobe and microprobe analysis. Hydrogen content increases with decreasing fO2 from 0.3 wt % at logfO2(IW) = -2.2 to 0.4 wt % at logfO2(IW) = -3.9. Nitrogen content increases with decreasing fO2 from 0.5 wt% at logfO2(IW)= -2.2 to 1.9 wt % at logfO2(IW)= -3.9. 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 a magma ocean, under fO2 conditions relevant to those prevailing during metal segregation.

The experimental results in the system model silicate melt (NaAlSi3O8 80% wt +FeO 20% wt) + molten Fe metallic phase + C+ H conducted at 4 GPa and 1550°C and logfO2 from 2 to 4 logfO2 (IW) allow to assume that the formation of compounds with C-H-type bonds (e.g., H2, CH4 and other molecules with such a bond) should be expected in primary melts of the reduced mantle, together with oxidized H species (as the OH-group). The relationship between them substantially depends on fO2.

We assume that the magmatic transport and chemical evolution of nitrogen, carbon and hydrogen during the reduced episode of early mantle evolution could be very much influenced by low fO2 values in presence of the metallic Fe phase. The primary melting is a way of providing the formation of the reduced forms of nitrogen, carbon and hydrogen (H2, CH4, NH3 together with H2O, OH) in magmas of the early Earth.

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