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Magmatic transfer of nitrogen carbon and hydrogen from reduced mantle during metal segregation

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In the theories of the Earth's formation the composition of gases extracted by primary planetary magmas is accounted for by a large-scale melting of the early mantle under fO_2 conditions relevant to those prevailing during metal segregation. In the framework of these concepts the gas regime of the early Earth should be related to dissolution features of volatile components in magmatic melts and melted Fe phase at low oxygen fugacity (fO_2) values lying below the fO_2 of iron-wüstite (IW) buffer equilibrium.

In a series of experiments in the system Fe-bearing melt + molten Fe phase + N + H + C conducted at 4 GPa, 1550 C, and $\log fO_2$ from 2 to 4 below $\log fO_2$ (IW) we have characterized the nature and quantified the abundance of N- H and C-compounds dissolved in a model silicate melt (Na_2O- FeO-Al_2O_3-SiO_2). . The infrared and Raman spectroscopy of glasses indicates a remarkable feature of [U+FFFD] N-H-O interaction with a reduced silicate: an appreciable change in the mechanism of their dissolution with a decrease in fO_2. The experimental results obtained at fO_2 falling in the stability region of the metallic Fe phase allow to assume that the formation of compounds with N–H-type and C–H bonds (CH_4), N–H–type bonds (NH_3, NH_4^+) and N_2 molecule should be expected in primary melts of the reduced mantle, together with oxidized H species (OH^-,H_2O). Some hydrogen is present in the melt in molecular form. Lowering fO_2 is characterized by an increase in the content of compounds with the C-H and the N–H– type bonds and a reduction of the content of oxidized hydrogen species, such as OH^- and H_2O.

The formation of N–H bonds in the reduced silicate melts results in a significant increase in nitrogen solubility that can reach 1-2 wt. %. Thus, hydrogen regime in depths of the mantle alongside with fO_2andpressure is apparently an essential factor of high nitrogen solubility in magmatic melts. The assumptions of probably higher nitrogen content in early melting products of the reduced Earth's mantle in comparison with nitrogen content in basalts of later geological time prove to be true (Libourel et al., 2003; Miyazaki et al., 2004; Roskosz et al., 2006; Mysen et al., 2008). Our researches have shown a critical role of hydrogen in this process.

Preliminary experimental data testify to a strong influence of pressure, oxygen fugacity, and hydrogen on distribution of nitrogen between liquid iron metallic phase and silicate liquid (D_N). It is found that at 4 GPa, 1550 ^oC, and $\Delta \lg fO_2$(IW)= -3.3 the value of D_N equals to 0.82, while at normal N_2 pressure, 1600 °C, and $fO_2 = fO_2$(IW) this value is much higher and close to 10^4. An essential decrease of D_N in our experiments is a direct result of growing nitrogen solubility in N-H silicate melt and Fe metallic phase at high pressure and the fO_2 values substantially below fO_2(IW).

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 fO_2values 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 (H_2, CH_4, NH_3 together with H_2O, OH) in magmas of the early Earth.

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