Metal-silicate melt partitioning of Ni with ab initio simulation methods

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Element partitioning plays a crucial role in understanding and explaining processes during the formation and early evolution of the Earth or impact events. In recent years, the advance of computational methods and technology has made it possible to use ab initio molecular simulations as a complementary tool to experimental approaches. Here, we present a pilot study to predict Ni partitioning between metal and silicate melts at 2500 K and ambient pressure. Using the alchemical transmutation method of ab initio thermodynamic integration and the CP2K program package, we compute free energy changes and describe exchange reactions of Ni between silicate and metal melt.

The melt compositions for the model systems were chosen to represent experimentally determined equilibrium compositions[1]. For the simulations, the silicate melt of 112 atoms contains only one Ni atom (approx. 2 wt%). The metal phase is reproduced by a mixture of 48 Fe and 16 Ni atoms. With an equilibrium constant of K=1.05, the calculations confirm that under these conditions and compositions, the reaction

$$Fe^{2+}_{silicate} + Ni_{metal} \rightarrow Fe_{metal} + Ni^{2+}_{silicate} \qquad (1)$$

is close to equilibrium. A partition coefficient of D_{Ni}=450±100 is estimated based on thermodynamic integration results, which is very close to the experimental value (D_{Ni} =397+31-28[2]).

Changes in the Ni contents of both metal and silicate melt indicate that the amount of Ni in the silicate melt is the crucial factor that determines the partitioning.

This behavior can be traced back to structural effects in the silicate melt. The Ni-O bonds are considerably longer than the Fe-O bonds, which may lead to structural distortions when larger amounts of Ni are incorporated. Thus only little Ni will be transferred to the silicate. On the other hand, in the metal melt the coordination environment of Ni and Fe is practically the same and it can take up a significant amount of Ni.

[1] Bouhifd and Jephcoat (2011) *Earth Planet. Sci. Lett.* **307**, 341-348. [2] Thibault and Walter (1995) *GCA* **59**, 991-1002.