Nickel isotopic fractionation between metal and silicate: insights from theoretical approaches

M.Méheut[1], D. Connétable[2], J. Guignard[3] and G. Quitté[3]

[1]GET, CNRS, Université de Toulouse, UPS, IRD, CNES, Toulouse, FRANCE
[2]CIRIMAT, INP , Toulouse, FRANCE
[3]IRAP, CNRS, Université de Toulouse, UPS, CNES, Toulouse, FRANCE

Ni isotopes cover a wide range of composition in meteorites that could trace different conditions of metal-silicate differentiation. In this work, we have computed the fractionation of Ni isotopes at equilibrium between metal and various silicates (Ni-olivine, Ni-diopside, Ni-talc), considering only the variation in vibrational free energy upon mass change. These computations are based on first-principles approaches, and rely on the Generalized Gradient Approximation to Density Functional Theory (GGA-DFT).

So far, calculations of Ni isotope fractionation have only concerned dissolved species (Fujii et al 2015), and this is the first time that metals and insulator solids are considered. Electronic structure computation of phases containing transition elements such as Ni present specific difficulties, first because of the magnetic nature of these phases, and second because of the failure of the GGA-DFT approach to take into account the insulating nature of Ni-silicates. As a consequence, several theoretical frameworks were tested for these phases (several possible magnetic orders, with or without DFT+U correction). We conclude that, if a proper modeling of magnetic order may have a significant impact (on the order of 20% of the overall olivine-metal fractionation), a simplified computation appears sufficient to semi-quantitatively estimate these properties.

At 1000K, we find a fractionation of -0.016, -0.014, and -0.042‰.amu^{-1} between pure Ni metal and Ni-olivine, Ni-diopside and Ni-talc, respectively. The temperature dependence is essentially linear in $10^6/T^2$. The very sign of this metal-silicate fractionation is in contradiction with the experimental findings of Lazaar et al 2012 (Ni-metal/Ni-talc = +0.063‰.amu^{-1}), but consistent with the results of Guignard et al 2017, which showed no measurable Ni isotopes equilibrium fractionation between silicate melt and Ni metal at high temperature (1623 K).

Reasons for this discrepancy, and the specificities of this isotopic system will be further discussed.

References
Fujii et al 2014 GCA V140, pp553-576
Lazar et al 2012 GCA V 86 pp 276-295
Guignard et al 2017, Goldschmidt abstract