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Experimental investigation of metal-silicate Germanium isotopic fractionation at equilibrium: insights into early planetary differentiation

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The process of primitive differentiation and the formation of metallic cores are crucial stages in the evolution of terrestrial planets, the Moon, and asteroids. To better understand this process, specific geochemical tracers such as germanium (Ge) are required. Its moderate siderophile and volatile nature is highly dependent on temperature, pressure, and fO_2 conditions [1-3]. Significant variations in Ge isotopes between silicate and metallic phases have been observed on different scales (chondrites, Fe-meteorites, and planetary silicate reservoirs [4-6]), identifying a positive metal-silicate mass-dependent isotopic fractionation of Ge.

To quantify metal-silicate Ge fractionation at equilibrium, time-series experiments (from 20mn to 168h) were performed using a piston cylinder apparatus (CRPG-Nancy), with Fe-Ni capsules and »3200 ppm Ge-doped CMAS silicate starting materials, at 1350°C, fO_2»IW-2.5, and 1GPa to prevent Ge evaporation. Both hand-separated metal and silicate phases were analyzed for Ge using a combination of bulk (HG-MC-ICP-MS) [4] and in-situ (LA-ICP-MS) techniques. High-resolution (»10µm) multi-element spectral imaging (LIBS) provided a novel and powerful approach for interpreting chemical and isotopic data.

The LIBS images revealed that the existence of Ge-rich silicate crystals in some experiments, regardless of duration, leads to a decrease in the bulk Ge content of the solid Fe-Ni capsule, suggesting a preferential compatible behavior of Ge. Preliminary results, in crystal-free experiments, show an increase of Ge content in the metal phase overtime (20mn to 48h), ranging from 307 to 1380 ppm, and of $\delta^{74}Ge_{metal}$, lighter than the CMAS composition, from -5.90±0.03‰ to -1.89±0.08‰ (2σSD), with an isotopic equilibrium attained after 3h. In such closed system, we expect $\delta^{74}Ge_{silicate} > \delta^{74}Ge_{metal}$, thus a negative $\Delta^{74}Ge_{metal\text{-}silicate}$, as opposed to natural sample observations [4-6]. Forthcoming results on the silicate phase will allow to propose an additional kinetic isotopic fractionation occurring during Ge loss through volatility as planetesimals and planets formed.

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