

Primary Metal-Silicate Differentiation of Planetesimals: Isotopic Fractionation of Germanium in Iron Meteorites and in the Earth Crust

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Mantle-core differentiation processes have been studied through the partition of siderophile elements (Mo, W, Co, Ni) in the earth mantle. Contents of these siderophile elements in the silicate mantle of the Earth seem to be in excess compared to theoretical calculation of mantle-core equilibrium processes. Models based on experimental studies of metal-silicate partition coefficients (Schmitt et al., 1989) have invoked variations in oxygen fugacity from reduced to more oxidizing conditions at the end of accretion.

We present here another approach consisting of studying the isotopic behavior of a moderately siderophile element, Ge, in iron meteorites. Ge is one the key elements, with Ni, for iron meteorites classification, with a variation of an order of four in concentration, i.e. from 0.03-1.1 ppm to 520 ppm. All the Ge isotopes (masses 70,72,73,74,76) are stable and are not produced by any radioactive decay. Temperature and oxygen fugacity can produce potential conditions for isotopic fractionation. Ge^{4+} is the more common state, but under reduced conditions, the unstable and volatile divalent species GeO (Ge^{2+}) (Schmitt et al., 1989) can fractionate isotopically. With the assumption that iron meteorites can represent core of planets, the understanding of isotopic and elemental variations of Ge in iron meteorites can bring informations on the physicochemical conditions of Ge metal-silicate distribution. Ge isotopic analyses have been performed on the Hexapole Plasma Source Mass Spectrometry (Isoprobe, Micromass, CRPG-Nancy). The advantage of the Isoprobe compared to the Plasma 54 (VG Instruments) (Hirata, 1997) is due to the presence of a collision cell "Hexapole" which, with an added $\text{Ar}+\text{H}$ flux (Ar/H of 4.8-5.1 for Ge analyses), can reduce the spread in energy of ions entering in the magnetic sector, and neutralize ions with a high ionization energy. This is of significant importance for Ge isotopic analyses as Ar_2 , $^{38}\text{Ar}^{36}\text{Ar}$ and $^{40}\text{Ar}^{36}\text{Ar}$, interfere with Ge on 74 and 76 masses, respectively. External mass bias fractionation correction of Ge isotopic values with the international Ga isotopic standard SRM 994 ($^{69}\text{Ga}/^{71}\text{Ga} = 1.50676$) is done using exponential law. Isotopic analyses of Ge are carried out by adding 100 ppb of SRM Ga standard to a 1 ppm dilution of the JMC commercial solution of 1 000 ppm Ge standard. Internal

precision is of 0.01‰ and 0.02‰ on $^{70}\text{Ge}/^{73}\text{Ge}$, $^{72}\text{Ge}/^{73}\text{Ge}$, $^{74}\text{Ge}/^{73}\text{Ge}$, with an external reproducibility of 0.2‰, 0.05‰ and 0.08‰, respectively.

Ge isotopic analyses have been performed on four samples including one iron meteorite from the Ge-enriched group IAB: Magura (486 ppm), the anomalously enriched Butler meteorite (IRUNG) (= 2000 ppm), one terrestrial sample: a sphalerite (ZnS) from St-Salvy mine (France) with 200-300 ppm Ge, and an additional Aldrich Ge standard. Samples have been digested in screw-top Teflon vials using concentrated HNO_3 at 55 C. Ge is separated from the Fe, Ni, Zn, Co, Ga matrix on AGV 50X8 cation-exchange resin using HNO_3 0.5N. Perfect separation is necessary because of isobaric interferences species as FeO and NiO at masses 70, 72, and 74, 76, respectively, and Zn interferences at mass 70. Ge from the samples must be Ga-free, because of mass bias external correction with Ga standard. Magura sample has been used as a test for external reproducibility, and separate chemistry has been performed on five fractions. Results are presented in term of δ per‰ with respect to the JMC Ge standard, and are shown in Figure 1 $\delta(^{72}\text{Ge}/^{73}\text{Ge})$ vs $\delta(^{70}\text{Ge}/^{73}\text{Ge})$ below.

A significant fractionation of 6‰ in Ge isotopes between these samples is demonstrated, considering an external reproducibility on Magura sample of $0.28 (\pm 2\sigma/n)$. Iron meteorite samples have negative $\delta(^{72}\text{Ge}/^{73}\text{Ge})$ and $\delta(^{70}\text{Ge}/^{73}\text{Ge})$, and terrestrial samples have positive δ Ge values. The extent of isotopic variation for terrestrial samples is more important (3‰) than previously reported by Hirata (1997). Two important points can emerged: - except Butler meteorite, samples lie on a mass fractionation line, suggested that these positive and negative fractionations can be produced by processes of core formation or mantle-crust separation during specific conditions of oxygen fugacities and temperature, pressure to be determined, - Butler meteorite with the highest Ge content shows the strongest negative isotopic fractionation which does not follow the mass fractionation line. This raises the question of initial heterogeneity of the parent body.

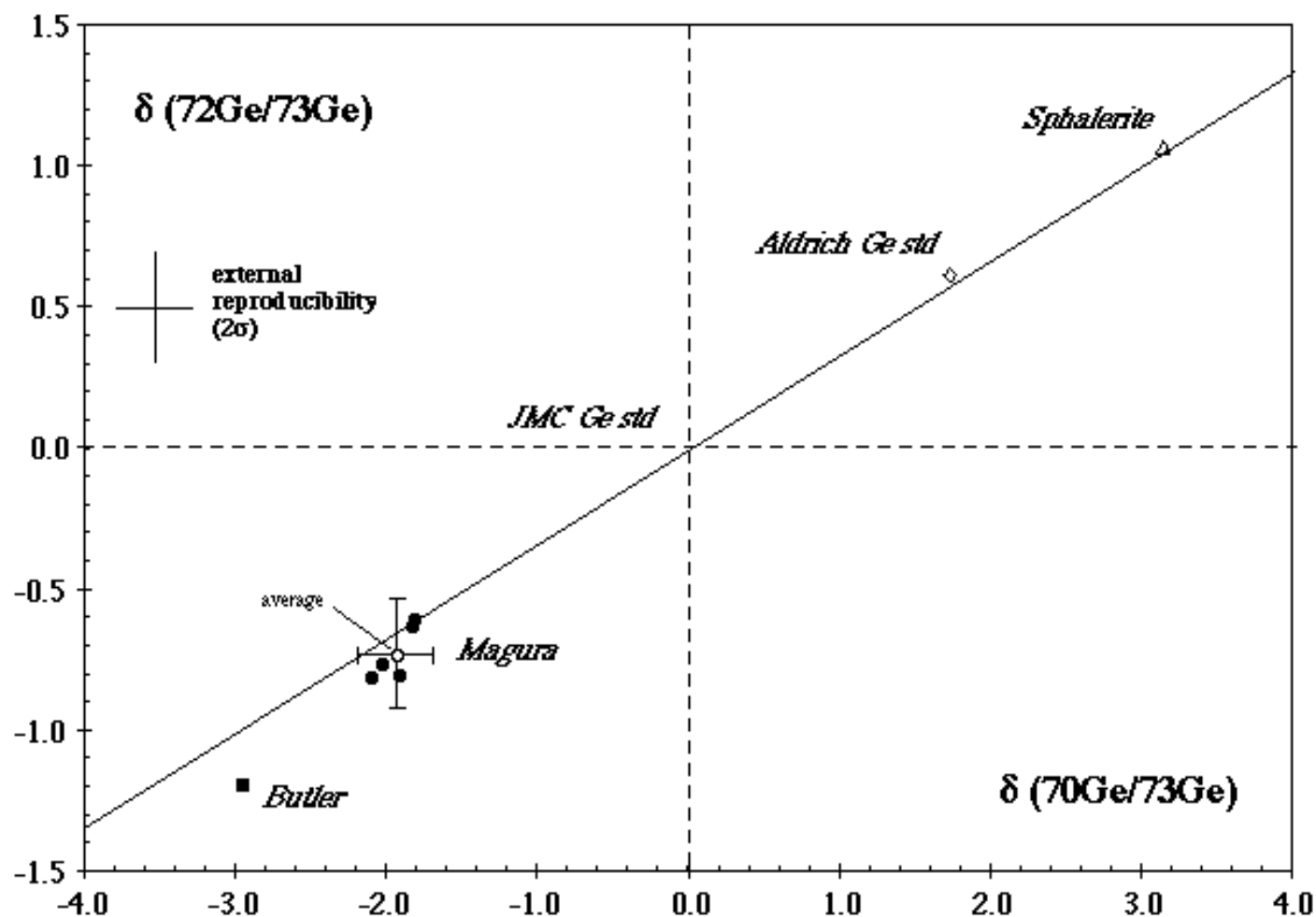


Figure 1: $\delta(^{72}\text{Ge}/^{73}\text{Ge})$ vs. $\delta(^{70}\text{Ge}/^{73}\text{Ge})$ for iron meteorites and two terrestrial samples (a sphalerite and a Aldrich Ge standard). External reproducibility (2σ) are given on Magura sample. The line drawn corresponds to mass-dependent fractionation line with a slope of 1/3.

Schmitt W, Palme H & Wanke H, *Geochim. Cosmochim. Acta*, **53**, 173-185, (1989).

Hirata T, *Geochim. Cosmochim. Acta*, **61**, 4439-4448, (1997).