

### Equilibrium iron isotope fractionation in metal sulfidation experiments at 600–1200 °C

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**Introduction:** Iron metal and troilite co-exist abundantly in many meteorite groups, including ordinary chondrites and iron meteorites [1,2]. Stable isotope systematics between metal and sulfide phases are affected by kinetic effects during cooling and phase separation between immiscible Fe and FeS from an initially homogeneous Fe-S melt. The equilibrium stable isotope composition of Fe between metal and troilite is therefore difficult to obtain from natural materials.

**Methods:** Here, we present results from Fe metal equilibrium sulfidation experiments in evacuated silica glass tubes at 600, 800, 900, 1000, 1100, and 1200 °C. Iron metal pellets were placed in graphite cups that were loaded in SiO<sub>2</sub> glass ampoules with elemental S at a molar ratio of Fe/S = 4. The glass ampoules were evacuated to  $\sim 10^{-5}$  bar and sealed with a H<sub>2</sub>-O<sub>2</sub> flame. The glass ampoules ( $\sim 4$ cm long) were suspended in the hot zone of a vertical tube furnace for up to 260 h. The experiments were mounted in epoxy resin and imaged by SEM. The two distinct phases, Fe and FeS, were analyzed for their  $\delta^{56}\text{Fe}$  and  $\delta^{57}\text{Fe}$  compositions with a high mass resolution MC-ICP-MS (Thermo Scientific Neptune Plus). The materials were ablated using a Spectra-Physics Solstice femtosecond laser ablation system at the institute of Mineralogy, Leibniz University Hannover, see [3] for analytical details.

**Results:** At 600–1000 °C the sulfidation reaction occurred in the solid state (Fig. 1 a, b), whereas at 1100 and 1200 °C we observe Fe and FeS melts (Fig. 1 c, d). Below the liquidus the Fe pellets are coated with FeS, and we observe minor porosity at the interface. Above the liquidus the melts mixed and coalesced, obscuring the initial pellet-coating relationship from the experimental setup. We insured isotopic equilibration by running experiments with different durations. At 600 °C and 71h we observed kinetic fractionation effects, whereas after 260h the sulfide coating was isotopically homogeneous. In Fig. 2 we show the  $\delta^{56}\text{Fe}$  compositions of the Fe and FeS phases as a function of temperature, normalized to the isotopic composition of the starting material. Overall, FeS is systematically heavier compared to the Fe metal, and the overall isotopic fractionation decreases with increasing temperature from 600 to 1200 °C. However, a comparison of the experimental results with  $\delta^{56}\text{Fe}$  data from ordinary [1] and iron [2] meteorites shows the opposite fractionation trend. In the meteorites the FeS is always isotopically lighter compared to the Fe metal. We suggest that this is due to kinetic fractionation effects that occur during cooling of the Fe-FeS melts in the meteorites, whereas our quenched experiments retained the equilibrium  $\delta^{56}\text{Fe}$  fractionation.

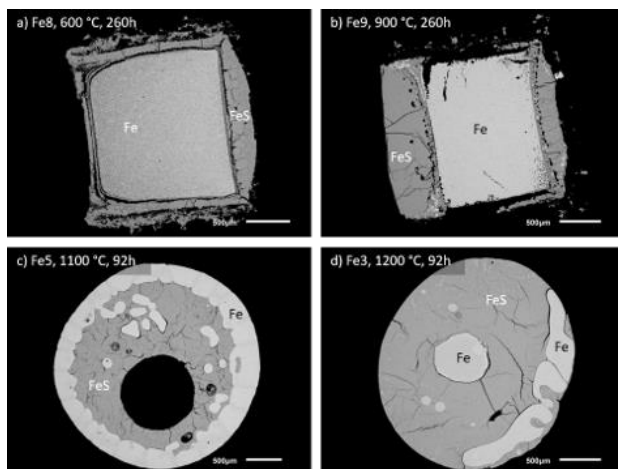


Fig. 1: Backscattered electron images of experiments at a) 600, b) 900, c) 1100, and d) 1200 °C. The Fe metal appears brighter in the image compared to the troilite. At 1100 and 1200 °C the experiments were molten.

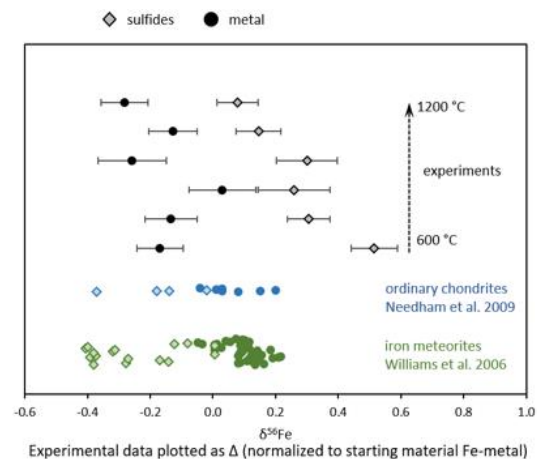


Fig. 2:  $\delta^{56}\text{Fe}$  compositions of the experimental Fe (circles) and FeS (diamonds) compared to Fe and FeS isotopic compositions in ordinary chondrites [1] and iron meteorites [2]. Note that in the experiments the sulfides have a heavier  $\delta^{56}\text{Fe}$  composition compared to the metal, whereas the isotopic relationship is inverse in the meteorites.

### References:

[1] Needham A. W. et al. (2009) *GCA* 73:7399–7413. [2] Williams H. M. et al. (2006) *EPSL* 250:486–500. [3] Horn I. (2007) *Spectrochim. Acta B* 62:410–422.