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Experimental and theoretical insights into the acquisition of kinetic iron isotope signatures of sedimentary pyrite

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The chemical and isotope composition of pyrite (FeS_2) is often used as a proxy of paleoredox conditions. However, the mechanisms by which isotopic signals are recorded in sedimentary pyrite are still incompletely understood, especially for iron isotopes. Here, we report iron isotope time series for pyrites that were experimentally produced through the polysulfide pathway at ambient temperature. The iron isotope solid-solution partitioning significantly departs from equilibrium as soon as pyrite nucleates and until the end of our experiments. We observe a strong inversion of the isotope partitioning between solid and solution, which coincides with the transformation of disordered mackinawite to pyrite. Regardless of the timing of this transformation, which can be controlled by the addition of As or Ni impurities (Baya et al. 2021), the final partitioning between pyrite and solution results in the same $\Delta^{57}Fe$ value of approximately -3.5 ‰. This fractionation factor is similar to that previously found in pyrite precipitation experiments through the H_2S pathway (Guilbaud et al., 2011). Based on newly computed *ab initio* equilibrium fractionation factors and previous X-ray absorption spectroscopic measurements (Baya et al. 2021), we propose that an isotopically light iron molecular intermediate leads to pyrite nucleation and thus explains the observed fractionation in both the so-called H_2S and polysulfide pathways (Poitrasson et al. 2026). This fractionation process may thus be common to both abiotic pyrite production pathways and could be thought as being at the origin of the lightest $\delta^{57}Fe$ isotope signatures found in the Archean pyrite sedimentary record.

References

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