

## **In situ Fe and S isotope composition of pyrites from the 3.24 Ga old Mendon Formation, South Africa**

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Archean sedimentary pyrites record Fe isotope variations through time, with particularly large negative excursion of  $\delta^{56}\text{Fe}$  values between 2.7 and 2.3 Ga. The origin of this negative excursion is strongly debated but was first interpreted as reflecting changes in seawater iron cycle related to the prelude of the Great Oxygenation Event. However, this view of the Fe cycle recorded in pyrite may have been biased by a lack of Fe isotope data for pyrites older than 2.7 Ga.

We report in situ  $\delta^{56}\text{Fe}$  and multiple S isotope compositions of pyrites from the 3.24 Ga old Mendon formation, Kaapvaal craton, South Africa. Pyrites from ten chert and altered komatiite samples from the Barberton Barite Drilling Project (BBDP) have been analysed. High-resolution TEM observations reveal that these pyrites are polycrystalline and contain micro and nanoscale mineral inclusions (quartz and carbonates) from the host-rock, implying a late diagenetic origin. In situ Fe and S isotope compositions of pyrites were measured with ims 1280 HR2 at CRPG (Nancy), with a reproducibility better than 0.2 ‰ ( $2\sigma$ ) for both  $\delta^{56}\text{Fe}$  and  $\delta^{34}\text{S}$  and 0.1 ‰ ( $2\sigma$ ) for  $\Delta^{33}\text{S}$ . Highly variable values of  $\delta^{56}\text{Fe}$  (-4.3 to +3.2‰),  $\delta^{34}\text{S}$  (-2.49 to +6.22‰) and  $\Delta^{33}\text{S}$  (-0.39 to 4.25‰) were obtained. Strikingly, the  $\delta^{56}\text{Fe}$  values deeply extend the range of values reported for archean pyrites. In addition, the  $\delta^{56}\text{Fe}$ - $\Delta^{33}\text{S}$  distributions display three different regions, which we interpret as a result of various pathways of pyrite formation from three different and uncorrelated sources of Fe and S. This in situ coupled Fe and S isotope study of 3.24 Ga pyrites thus shows an extreme isotopic variability for both Fe and S, at the micrometer scale, which highlights a complex diagenetic pathway of pyrite formation.

The preservation of a polycrystalline structure linked in space with  $\mu\text{m}$ -scale Fe and S isotope heterogeneities argues for a primary (early diagenetic) signature, which was not overprinted by secondary hydrothermal and metamorphic processes.