

***In situ* multiple sulfur isotope analysis of sedimentary pyrites with SHRIMP-SI: Unravelling complex depositional and post-depositional processes**

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The sulfur isotopic record of Archean and Paleoproterozoic sedimentary rocks places important constraints on the timing of atmospheric oxygenation. However, many of these ancient rocks have endured several post-depositional processes (e.g., diagenetic, magmatic, hydrothermal, and metamorphic) over geological time so that the original isotopic signature from the early atmosphere and biosphere is now largely overprinted.

In situ SHRIMP-SI measurements of multiple sulfur isotopes (³²S, ³³S, ³⁴S, ³⁶S) in pyrite now allow $\Delta^{33}\text{S}$ to be determined with internal errors better than 0.05‰ (2SE) and reproducibility about 0.1‰ (2SD). Charge mode measurements [1] of ³⁶S⁻ allow $\Delta^{36}\text{S}$ values to be determined with internal precisions of $\pm 0.2\%$ (2SE) and reproducibility better than 0.25‰ (2SD). This level of precision permits identification, at the micron scale, of preserved isotopic signatures of ancient atmospheric chemical and biological activity, as well as overprinted secondary processes.

Multiple sulfur isotope analyses carried out in pyrites from Archean and Paleoproterozoic sedimentary successions in Brazil (Jacobina Basin) and Australia (Turee Creek Group) provide isotopic evidence for multiple generations of pyrite, represented by distinct $\Delta^{33}\text{S}$ and $\Delta^{36}\text{S}$ signatures among individual pyrite crystals and within single grains with overgrowth textures. The level of precision and spatial resolution achievable with SHRIMP-SI even allows distinction of small but isotopically distinct pyrite populations that would otherwise not be resolved with bulk isotope data.

[1] Ireland *et al* (2014) *Int. J. of Mass Spect.* **359**, 26-37.