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Pyrite morphology evidence for highly fluctuating redox conditions in early Cambrian oceans

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The lower Cambrian black shales have been hypothesized to form under highly fluctuating redox conditions, which could account for the episodic preservation of early animal fossils within these anoxic deposits. However, direct evidence supporting this hypothesis has been lacking. In this study, we conducted a detailed morphological analysis of pyrites from the lower Cambrian Shuijingtuo (SJT) Formation in South China, utilizing Ar ion milling-scanning electron microscopy (SEM) and aberration-corrected transmission electron microscopy (AC-TEM) at micron to nanometer scales. Our investigation revealed framboidal grains with irregularly serrate margins in the upper SJT Formation, occurring in layers ranging from microns to centimeters in thickness. High-resolution scanning transmission electron microscopy (STEM) analysis demonstrated that some oxidation byproducts, including marcasite, ferrous sulfate, and iron sulfate hydrate are existed in these grains in the oxidized phase form. Furthermore, STEM elemental mapping identified a marginal ~5-nm-thick amorphous passivation layer composed primarily of Fe and O on some grains. We propose that these distinctive morphological characteristics of framboids resulted from partial oxidation and dissolution during syngenesis and early diagenesis, driven by high-frequency fluctuations in bottom-pore water redox conditions during the early Cambrian period. We suggest two stages of alteration were involved: Stage 1 involved the formation of a passivation layer on framboid surfaces during initial pyrite oxidation; Stage 2, along with increasing oxidation extent and elevated Eh in bottom waters or pore waters, the serrate structure was formed from the partial dissolution of pyrite framboids, and the precipitation of oxidation byproducts lead to a complex lattice structure. The highly fluctuating redox conditions inferred for the early Cambrian oceans are consistent with the large variability of MoEF (8.78~248.14), U/Th (0.54~24.59), and V/Cr (0.68~16.55) of studied black shales. Our findings provide direct pyrite-morphological evidence for the highly dynamic redox conditions in early Cambrian oceans, offering a plausible explanation for the unusual occurrences of early animal fossils in these anoxic black shales.