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The effect of iron sulfidation on the stability of the trisulfur radical ion $S_3^{\bullet-}$ in subduction zone fluids

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Sulfur cycling in subduction zones plays a critical role in the redox evolution of the mantle wedge and the formation of arc-related ore deposits. A growing number of studies indicate that, in addition to reduced sulfides (H_2S, HS^-) and oxidized sulfates (HSO_4^-, SO_4^{2-}), subduction zone fluids may also contain the stable trisulfur radical anion ($S_3^{\bullet-}$), which has been proposed as a dominant carrier of ore–forming elements in subduction zone fluids. However, existing high P–T experiments have primarily focused on $S_3^{\bullet-}$ stability in pure aqueous sulfide–sulfate systems. The role of iron sulfidation on S–rich fluids in subduction zone rocks, which may influence $S_3^{\bullet-}$ stability, remains poorly understood. In this study, we systematically investigated the impact of iron/iron oxides on $S_3^{\bullet-}$ stability in subduction-zone fluids using in situ high P–T experiments and thermodynamic simulations. Our results reveal that in pure aqueous systems containing both SO_4^{2-} and S^{2-}, $S_3^{\bullet-}$ is the dominant sulfur species at temperatures above 350 °C along typical subduction P–T paths. However, the introduction of Fe or FeO into these $S_3^{\bullet-}$–rich fluids triggers sulfidation reactions that lead to the precipitation of sulfide minerals (e.g., pyrite, pyrrhotite), substantially reducing $S_3^{\bullet-}$ stability in the fluid phase. In contrast, Fe_2O_3 has little effect on $S_3^{\bullet-}$ stability due to the formation of a passivating pyrite layer, which inhibits further sulfidation. Furthermore, we found that higher F/R mass ratios can mitigate the inhibitory effects of Fe and FeO on $S_3^{\bullet-}$ stability. We propose that $S_3^{\bullet-}$ in subduction zone fluids may serve as an ideal ligand for transporting sulfur–loving metals into the mantle wedge–provided that $S_3^{\bullet-}$–bearing fluids migrate through fluid channels with high F/R ratios and therefore minimal metasomatic effects on $S_3^{\bullet-}$ stability.