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Low aggregation state diminishes ferrihydrite reactivity

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Ferrihydrite is an abundant iron(oxy)hydroxide in soils and sediments and plays an important role in microbial iron cycling due to its high reactivity. Therefore, it is often synthesized and used in geomicrobiological and mineralogical studies. The reactivities of synthetic ferrihydrites vary between different studies and synthesis protocols. Hence, we synthesized five different ferrihydrites and characterized them with XRD, FTIR, XPS, and BET specific surface area. The reactivity of the ferrihydrite samples towards ascorbic acid was examined and compared with microbial reduction rates by Geobacter sulfurreducens.

FTIR and XRD results show the presence of secondary, higher crystalline iron oxide phases like goethite and akaganeite for two samples. Consequently, those samples revealed lower biotic and abiotic reduction rates compared to pure ferrihydrite. Comparison of reduction rates with the specific surface area of all ferrihydrites showed neither correlation with abiotic reductive dissolution nor with microbial reduction. Especially one sample, characterized by a very low aggregation state and presence of secondary minerals, revealed a poor reactivity. We speculate that apart from the occurring secondary minerals also the low aggregation state played an important role. Decreasing aggregation diminishes the amount of kinks and edges on the surfaces, which are produced at contact sites in aggregates. According to dissolution theories, dissolution mainly starts at those surface defects and slows down with decreasing amount of defects. Furthermore, the non-aggregated ferrihydrite is free of micropores, a further stimulant for dissolution. Independent repetitions of experiments and syntheses according to the same protocol but without formation of secondary minerals, confirmed the low reactivity of the non-aggregated ferrihydrite.

In summary, our results indicate that a decreasing aggregation state of ferrihydrite to a certain size does increase the reactivity. However, if the aggregation state decreases to the nanometer-scale, it does not obligatorily further increase the reactivity of ferrihydrite. Therefore, the general assumption that the reactivity of nanoparticles exceeds the reactivity of their macroparticulate counterparts should be verified for individual cases.